FEL photon sources and their multi-disciplinary applications

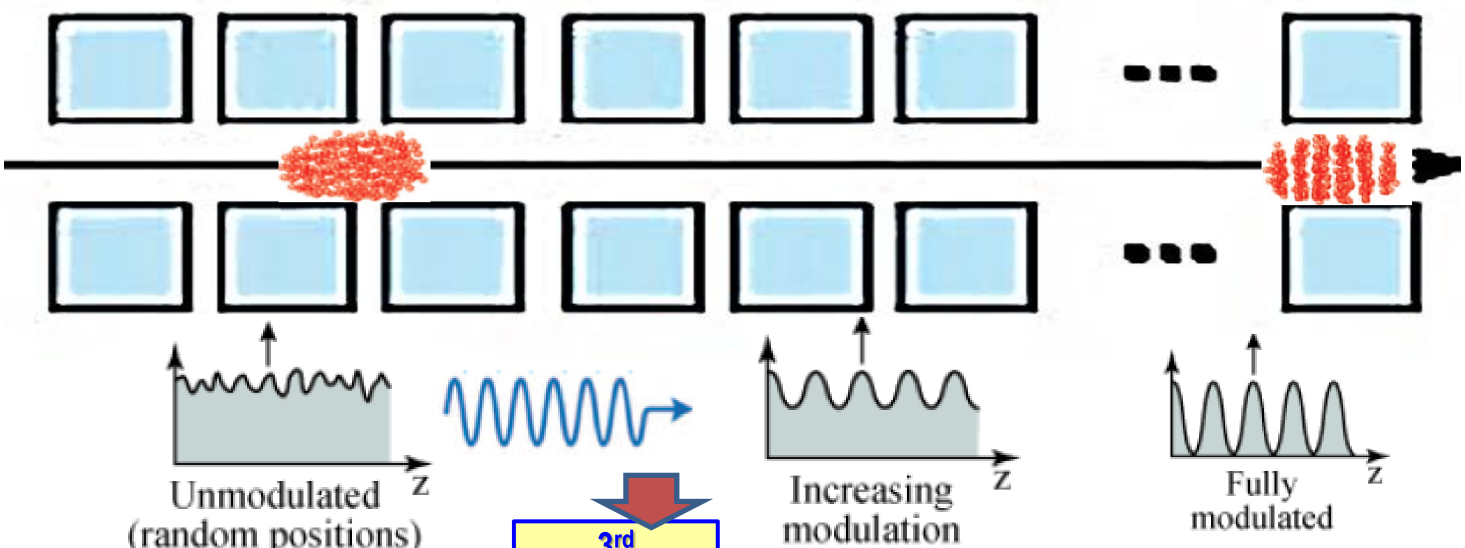
- EUV and X-ray FELs: brief notes on unique characteristics of FEL light.
- Selected application examples 'Static' and stroboscopic experiments using .
- What is next?

Maya Kiskinova

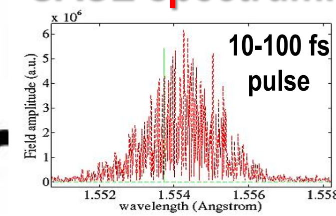
Elettra Sincrotrone Trieste

FEL PRODUCTION: Electron motion in a very long undulator evolves to a free electron laser

LINAC of electron bunches entering in a very long magnetic device the radiated fields become stronger and lead to microbunching, i.e. transform the random positions and motions of electrons into correlated waves of electrons, emitting radiation in phase.

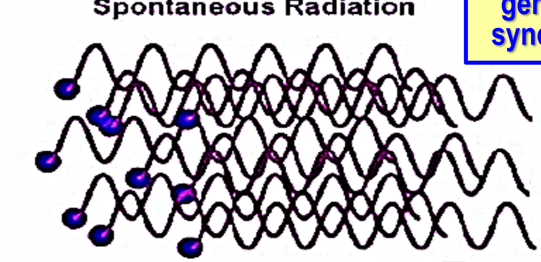


SASE spectrum.



envelope of a sub-pulses with random intensity, time duration, bandwidth

3rd generation synchrotron

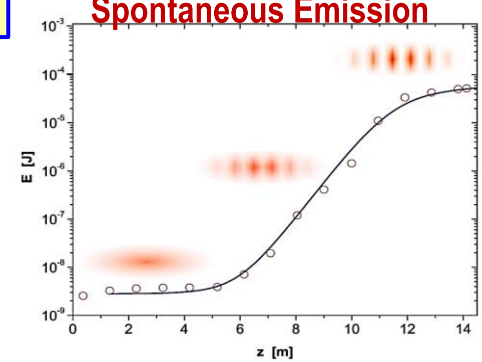


$E_{spt} \sim \sqrt{N}\, E_1$

$P_{spt} \sim N\, P_1$

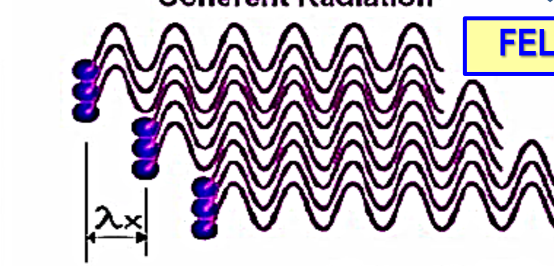
Uncorrelated radiated fields add limited coherence power N

Self Amplification of Spontaneous Emission



Courtesy of K-J. Kim

FEL



$E_{coherent} \sim N\, E_1$

$P_{coherent} \sim N^2 P_1$

Correlated radiated fields add - spatially coherent power N^2

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SASE-FEL Physics: increase of coherence power N as result of constructive interference of emitted radiation

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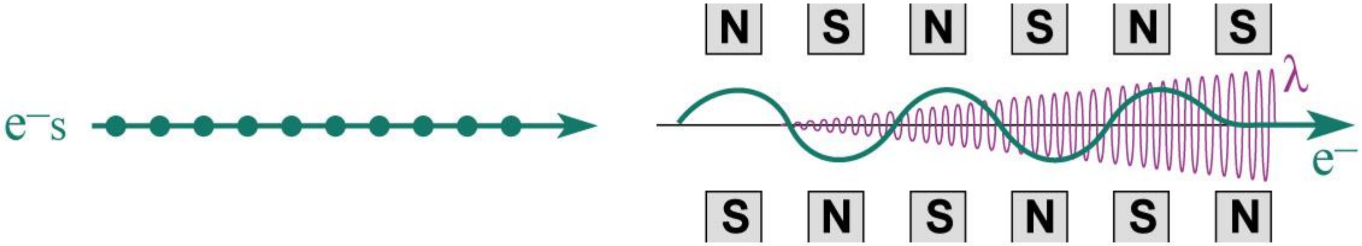
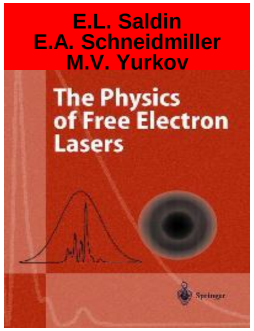
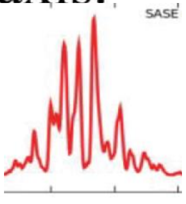


- High peak power;
- Very short fs pulses;
- High degree of spatial coherence



- Uniformly distributed particles (beam) into undulator.
- Emission of radiation ("spontaneous" emission).
- Wave grows enough (undulator radiation) to begin affecting. particle dynamics through m**a** = –e**E** radiation.
- Transverse coupling between $\mathbf{E}_{rad}$ and transverse velocity $\mathbf{v}_x$ (in undulator) leads to energy exchange between fields and particle (zero net at first) $\frac{dE_e}{dt} = mc^2 \frac{d\gamma}{dt} = \mathbf{F} \cdot \mathbf{v} = -e\, E \cdot \mathbf{v}_x$.
- Modulated velocities with increments in $\mathbf{v}_x$ lead to bunching on axis.
- Electron density modulation leads to stronger radiation, $P_{Tot} \propto \frac{Q^4}{M^2} \sim N^2 \frac{e^4}{m^2}$.

Time/energy structure: envelope of a series of sub-pulses with random intensity, time duration, bandwidth and phase.



- Stronger fields (wave) drive stronger transverse velocity.
- Stronger $\mathbf{v}_x$ drives stronger bunching, . . . stronger fields, . . . FEL action.

FEL_physics.ai

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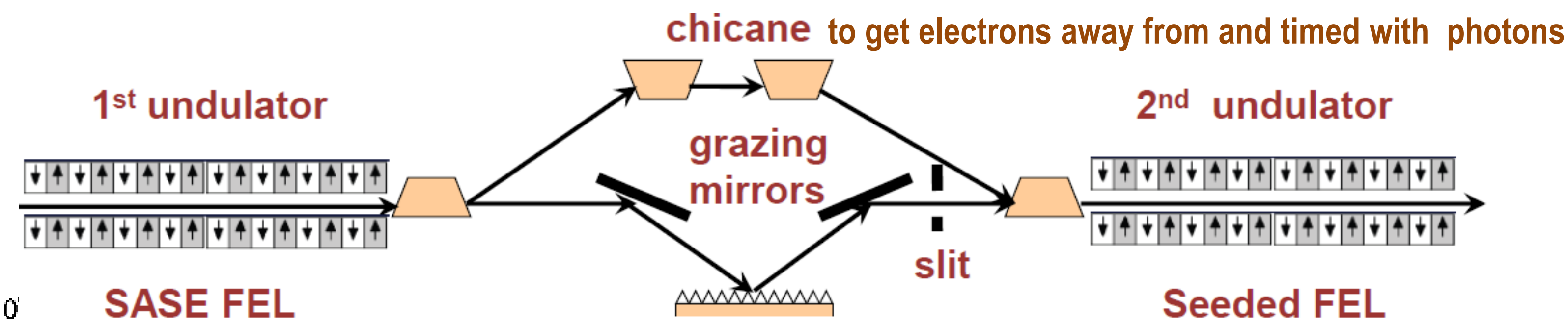
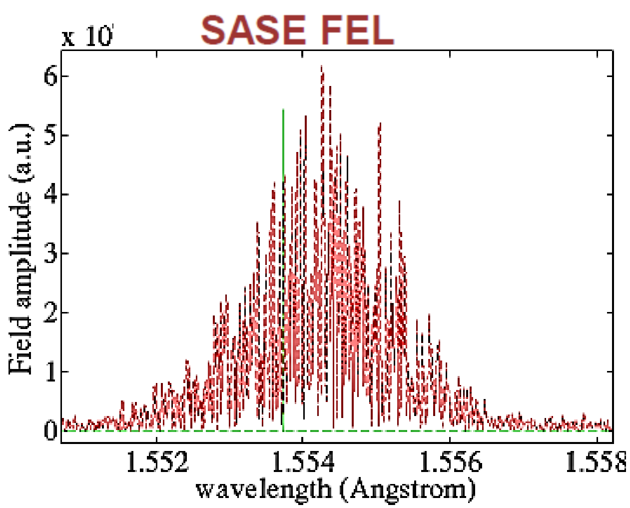
SASE-FEL: self-seeding for improving spectral (λ) purity

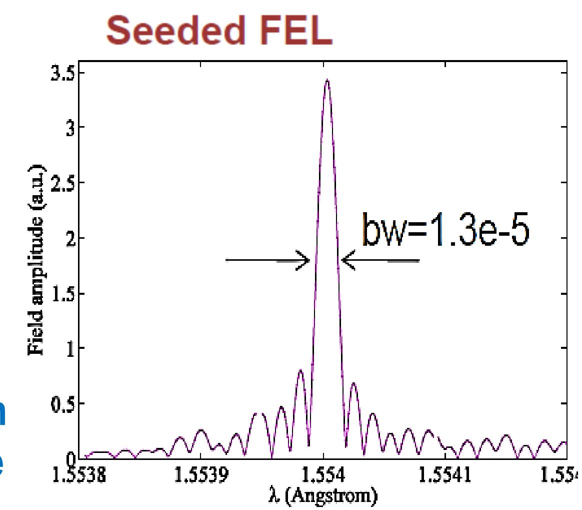
- First undulator generates SASE.
- X-ray monochromator (grating for soft X-rays or Si/diamond crystal for hard X-rays) filters SASE and generates seed.
- Chicane adds the same delays of electrons+washing SASE micro-bunching.
- Second undulator amplifies seed to saturation.





narrow bandwidth
enhanced peak power

Can use 2 bunches:
1st bunch generates seed light
2nd bunch timed to interact with seed light from 1st bunch in the second undulator



XIV School on Synchrotron Radiation: Fundamentals, Methods and Applications

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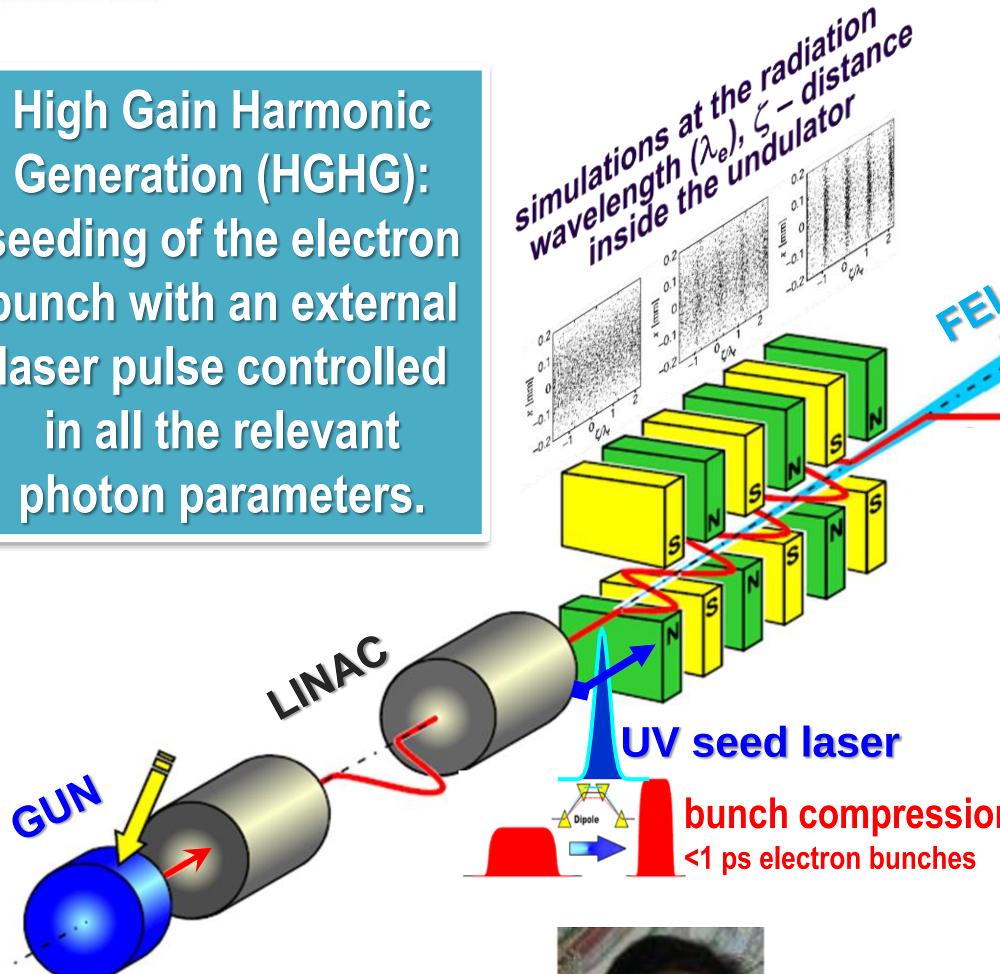
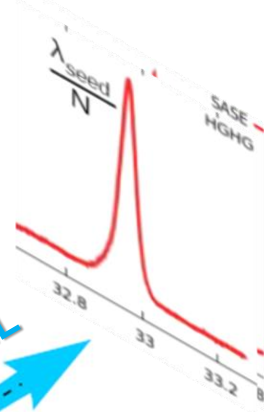
FERMI: Seeding SASE-FEL using optical laser

Elettra Sincrotrone Trieste

High Gain Harmonic Generation (HGHG): seeding of the electron bunch with an external laser pulse controlled in all the relevant photon parameters.

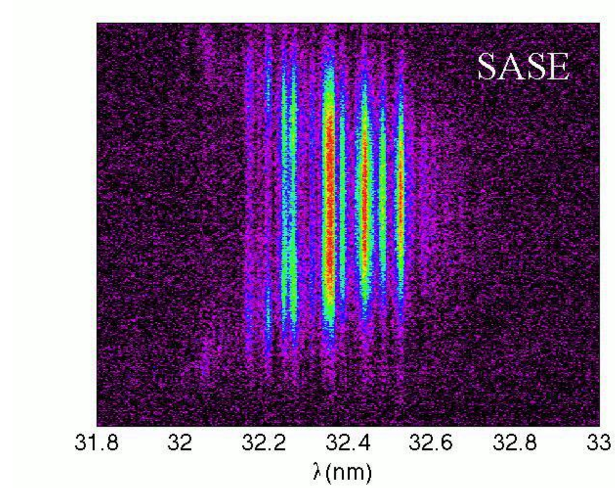




"SASE" FEL –time/energy structure of the pulse is an envelope of a series of sub-pulses with random intensity, bandwidth and phase.

The properties of the FERMI radiation are entangled with those of the seed laser.
Defined bandwidth-time profile.

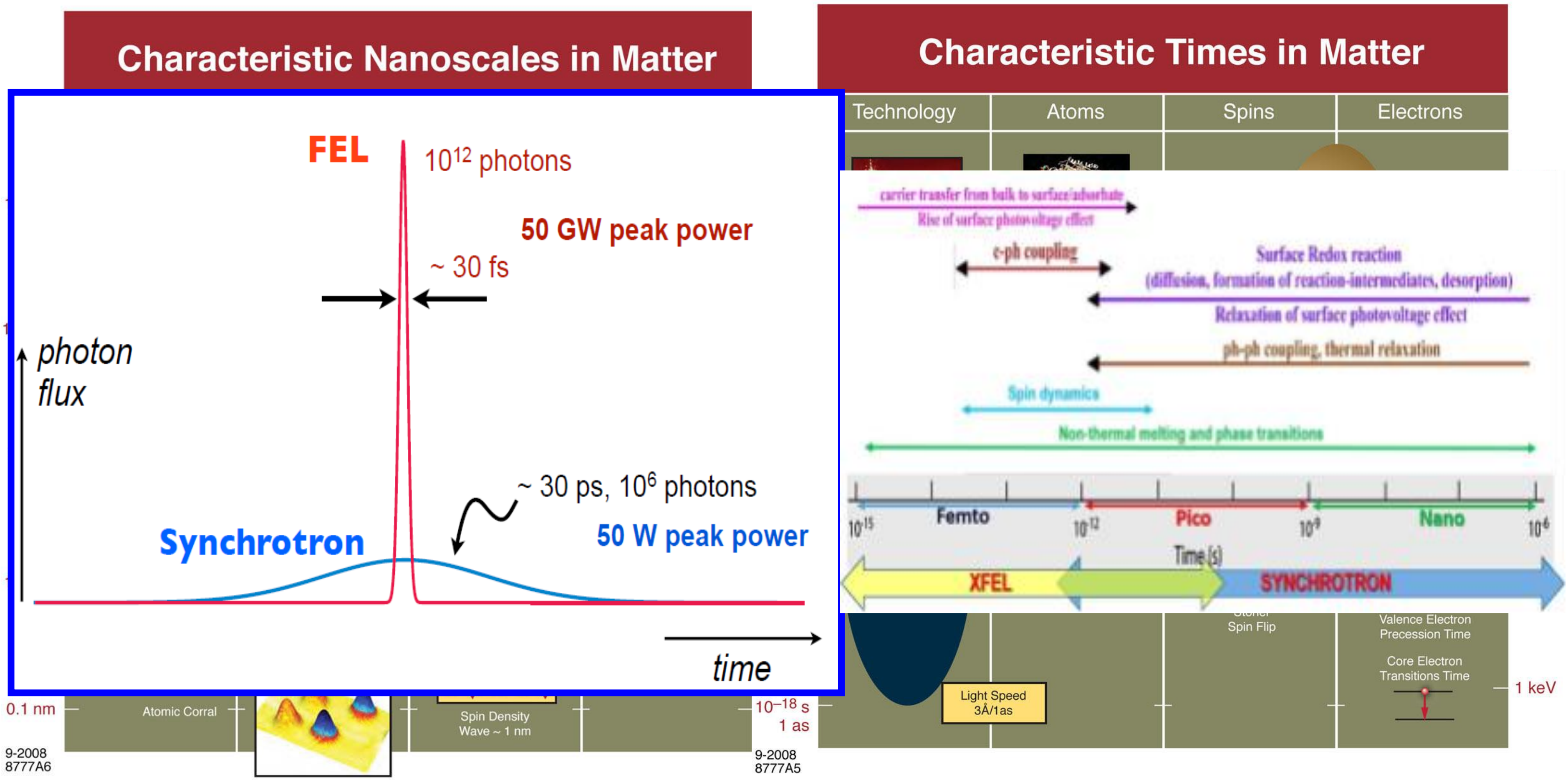
L. Giannessi



E. Allaria et al, Nat Photon. 6 (2012), 7 (2013)

Probing matter on nm length scales and and fs time scales

Unique new opportunities thanks to the distinct properties of FEL light: pulses with (i) High peak power, (ii) Ultrashort and (iii) Coherent



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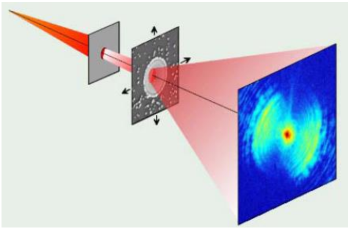
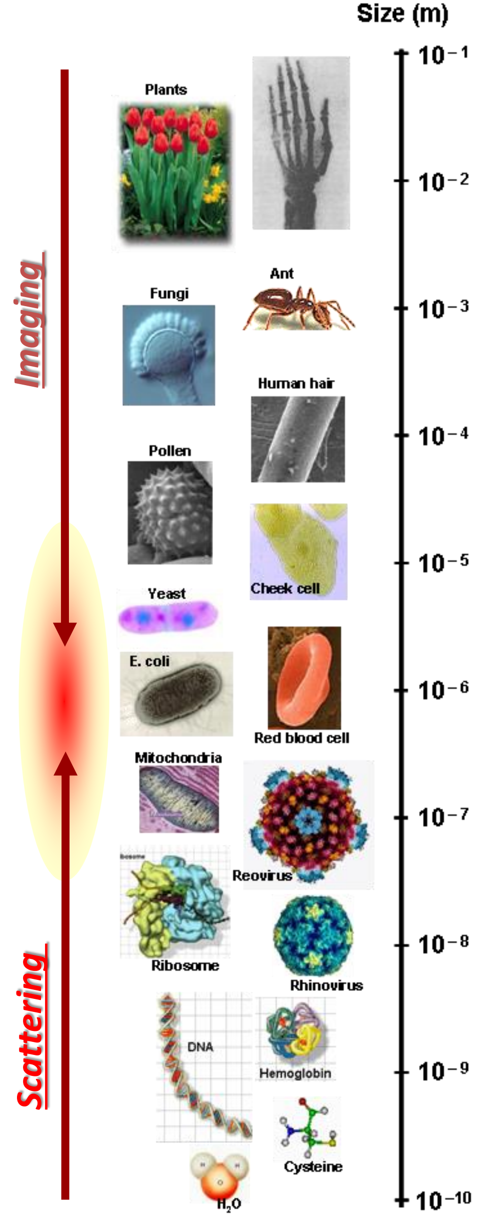
Imaging-resolution-penetration-time

- Scanning microscopes monitoring electrons - limited to surfaces.
- Transmission electron microscopes can resolve even atoms but are limited in penetration (samples thinner than ~ 30 nm).
- X-ray crystallography reveals the globally averaged 3D atomic structures based on the diffraction phenomenon, but requires crystals.
- Classical x-ray microscopy – limited in resolution and focal depth by the optical elements. Temporal resolution - ≥ ns

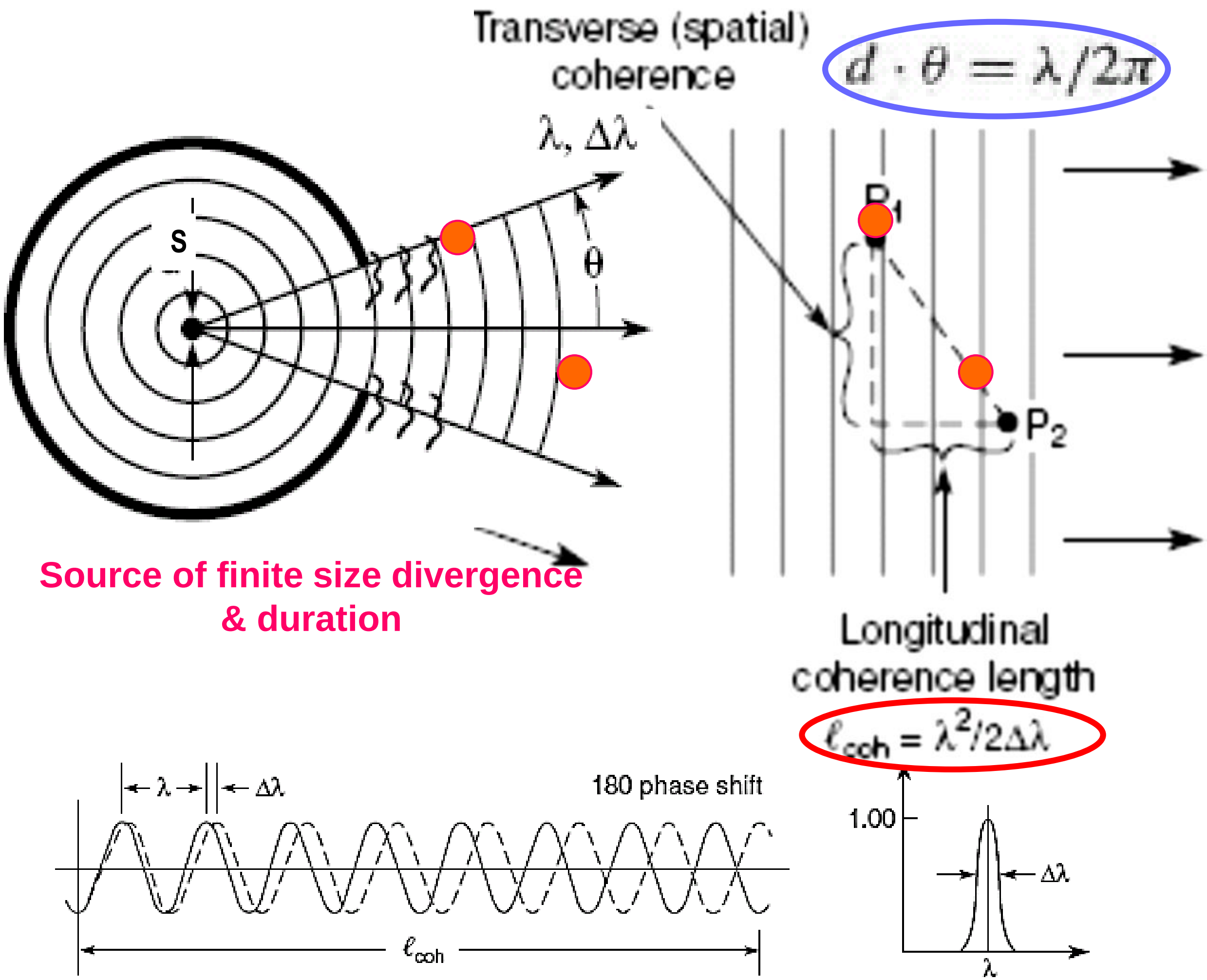
The optics depth and resolution limitations can be overcome by image reconstruction from measured coherent X ray scattering pattern visualizing the electron density of non-crystalline sample.





FEL: temporal and spatial coherence

A source with finite size and spectral bandwidth, restricted to radiate over a narrow solid angle, generates fields with strong phase and amplitude correlation to a limited extend

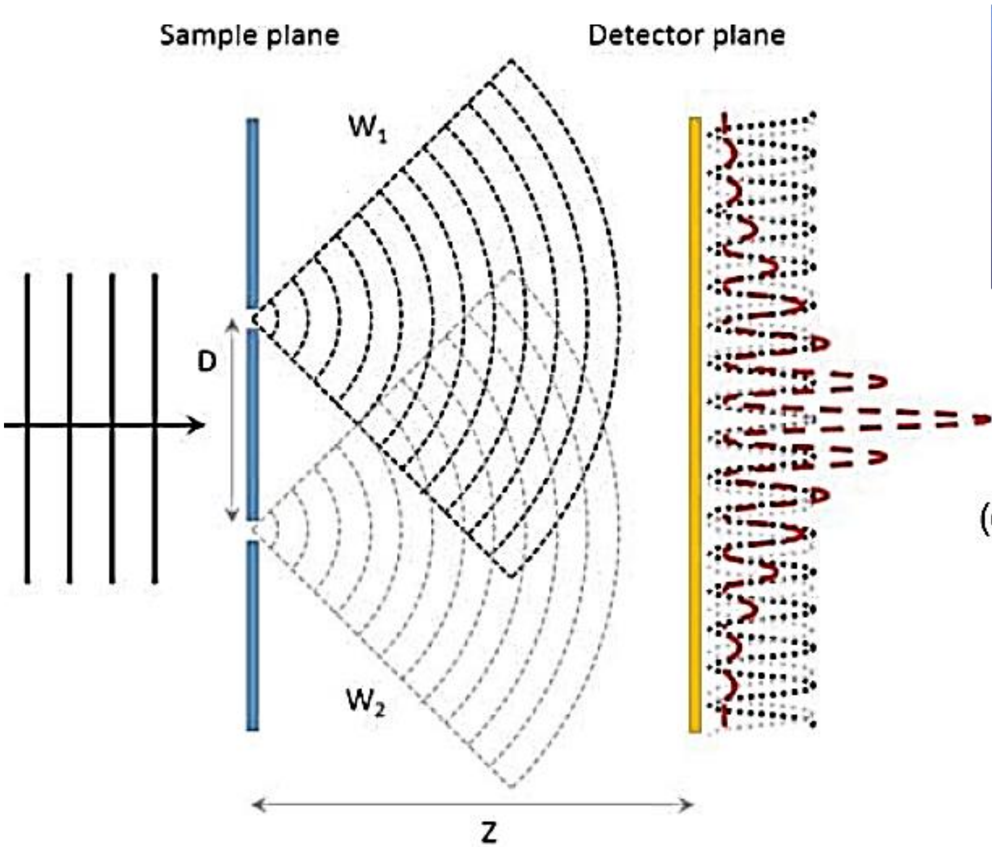


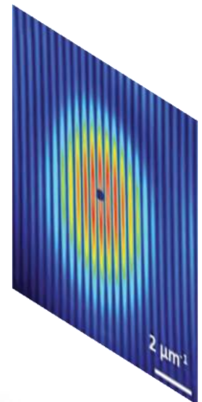
Transverse (spatial) Coherence:
depends on the source size, S, and angle of emission, Θ.

Longitudinal (temporal) Coherence:
depends on the finite spectral band width, Δλ/λ .

These requirements become very stringent for shorter wavelengths.

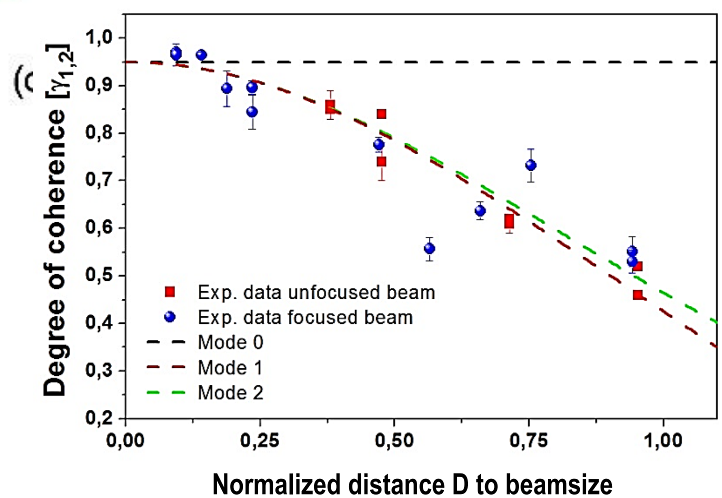
Young's double slit experiments as measure of spatial coherence

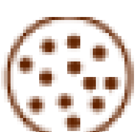




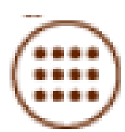
$$L^T_{coh} = \frac{2 \cdot \lambda \cdot Z}{\pi \cdot s}$$

the spatial distance along the two generated wavefronts out of phase by a factor $\pi/2$.





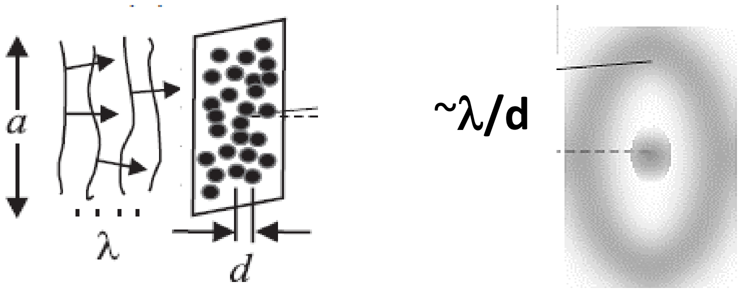
- Self-interference only
- Electric fields chaotic
- Intensities add



- Phase coherent electric fields
- Electric fields from all particles interfere constructively

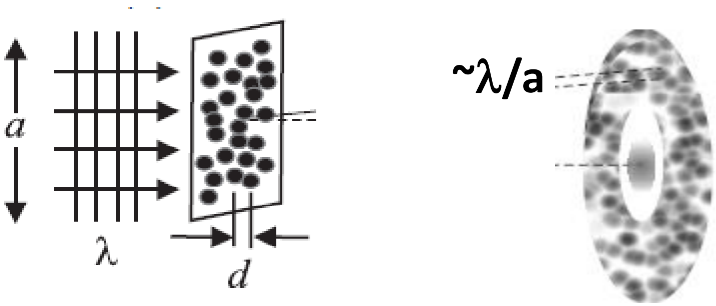
Coherent Diffraction Imaging (CDI) @FEL

based upon the principle of coherent scattering in combination with a method of direct phase recovery called oversampling



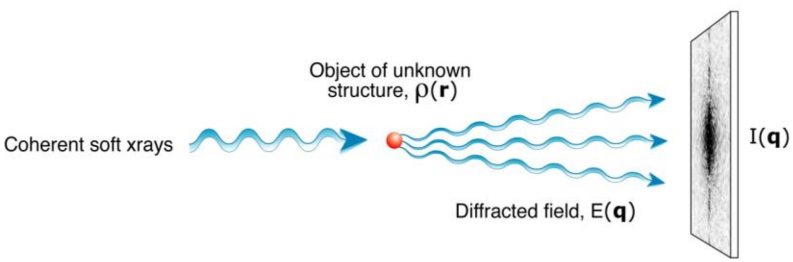
<u>Incoherent illumination:</u> coherence length smaller than the size, a, of sampled object

<u>Diffuse scattering</u> that averages over all features (sizes – correlations) resulting from slightly different wavefronts -



<u>Coherent illumination:</u> coherence length larger than the size , a, of sampled object

<u>Speckles encode exact arrangement</u> due to
interference of wavefronts scattered from the features - the positions of each feature, obtained by an iterative process inverting the pattern.



The scattered amplitude is Fourier transform of real space electron density f(r) of the object: $F(k) = \int f(r)\, e^{-2\pi i\, k \cdot r}\, dr$

- <u>Proposed by Sayre</u> to visualize the electron-density distribution in non-crystalline materials (1980)
- <u>Pioneering experiments: Kirz, Miao, Chapman, Spence, Robinson</u>, (Nature 400, 342; ibid. 442, 63; 448, 679; MRS Bull 29, 177, PNAS 102, 15343), *Science*, 316, 5830 etc)

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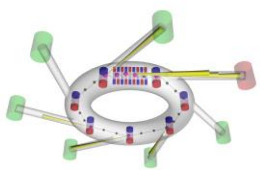
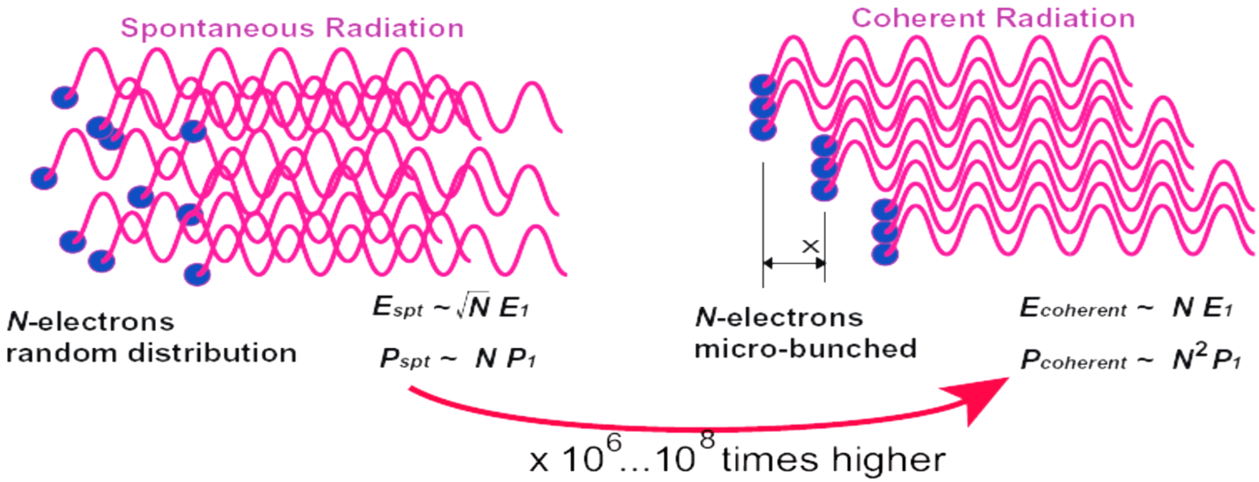
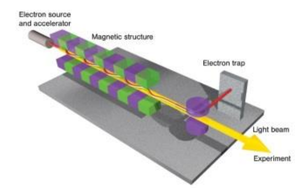
Coherent Diffraction Imaging:
Synchrotron vs FEL Radiation

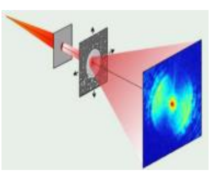
long pulses (sub-ns) max ~10^8 photons/pulse

Fs pulses > 10^{11} photons/pulse

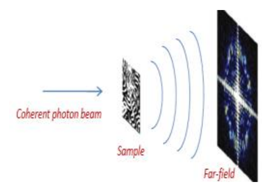






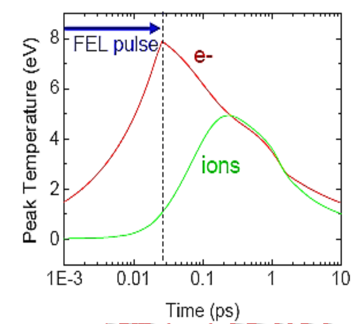


Synchrotron radiation:
pinhole and monochromators for spatial and spectral filtering, but at the expense of intensity!



FEL (FLASH, LSLS, SACLA, FERMI):
natural space coherence: each electron - spontaneous emission that overlap each other in phase

Ultra-short (fs) and ultra-bright coherent FEL pulses allow imaging with single pulse before the radiation damage manifests itself !



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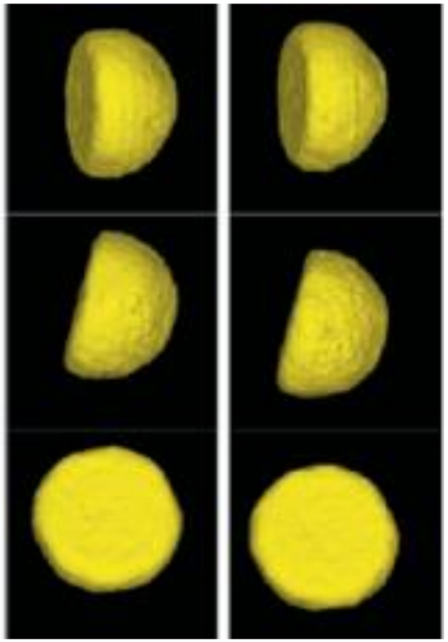
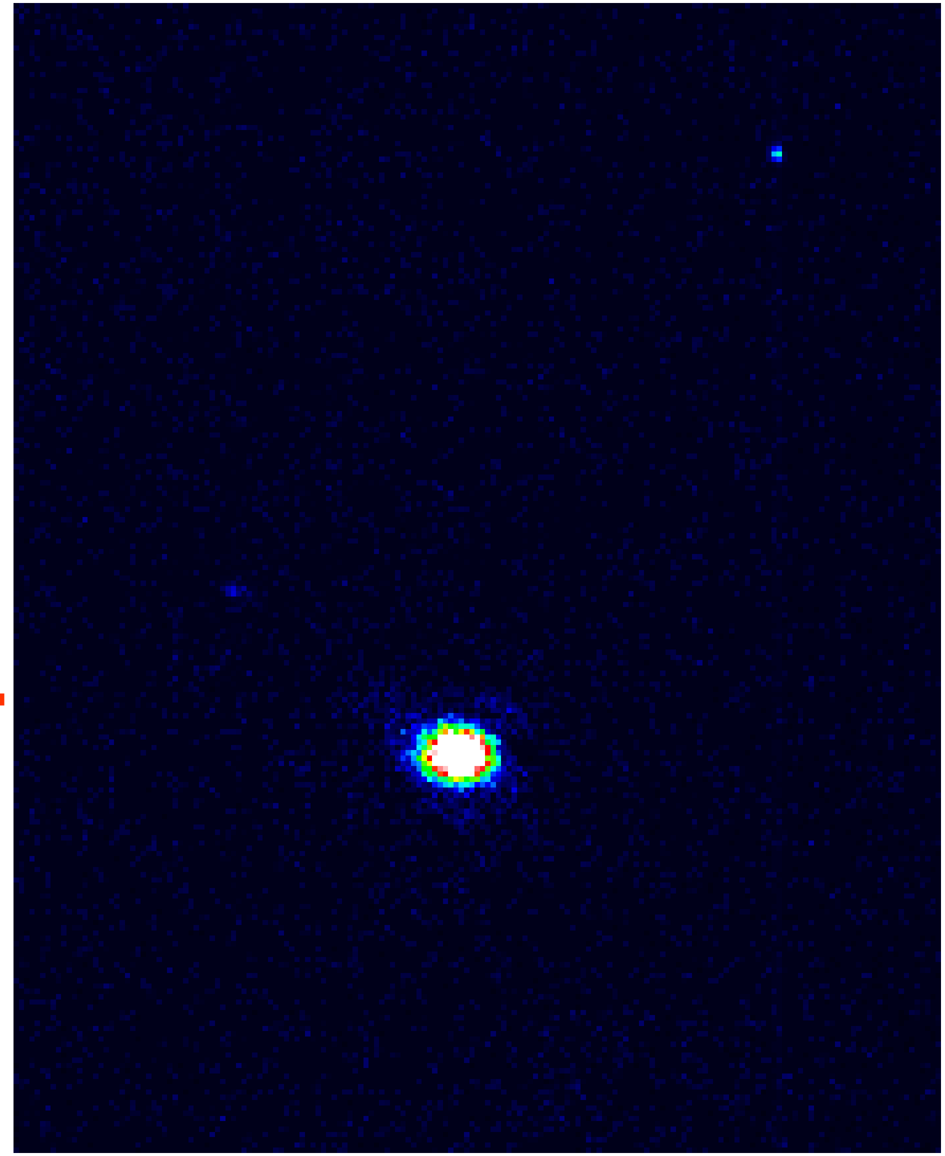
3D CDI of Ag cubes using synchrotron



Surface plots of reconstructed shapes



Rocking scan of Ag cubes with 0.01° steps,

courtesy K. Robinson, PRL 87, 195505

Resonant CDI: elemental and 'dichroic' sensitive

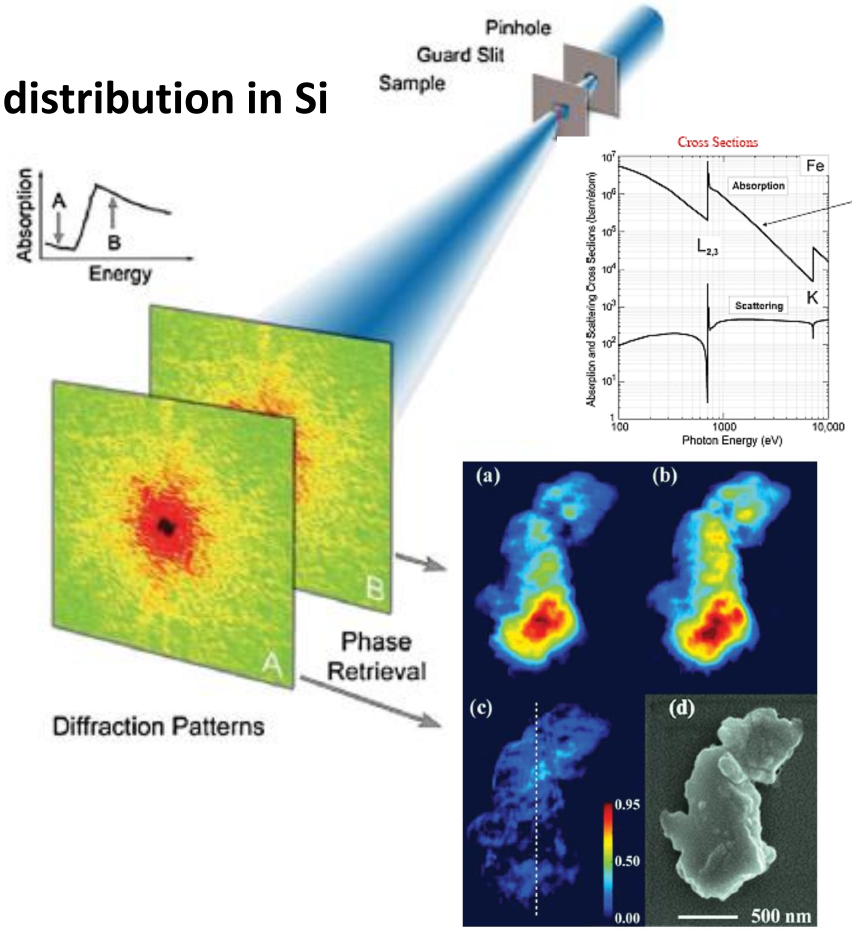
PRL 100, 025504 (2008) PHYSICAL REVIEW LETTERS week ending 18 JANUARY 2008

Nanoscale Imaging of Buried Structures with Elemental Specificity Using Resonant X-Ray Diffraction Microscopy

Changyong Song,[1] Raymond Bergstrom,[1] Damien Ramunno-Johnson,[1] Huaidong Jiang,[1] David Paterson,[2] Martin D. de Jonge,[3] Ian McNulty,[3] Jooyoung Lee,[4] Kang L. Wang,[4] and Jianwei Miao[1,*]

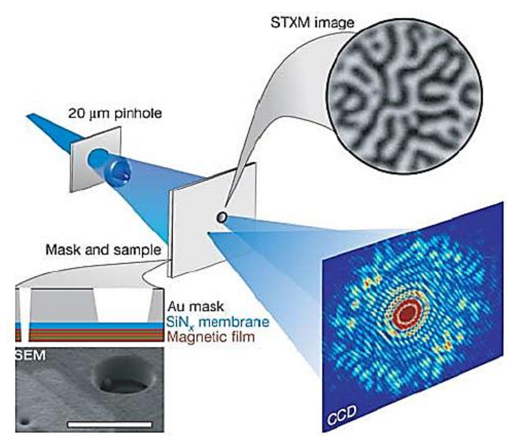
CDI is also sensitive to chemical states via near-edge resonances and can be extended to exploit other contrast mechanisms depending on resonant transitions such as x-ray magnetic circular dichroism. electronic orbital as well as chemical state.

Bi distribution in Si



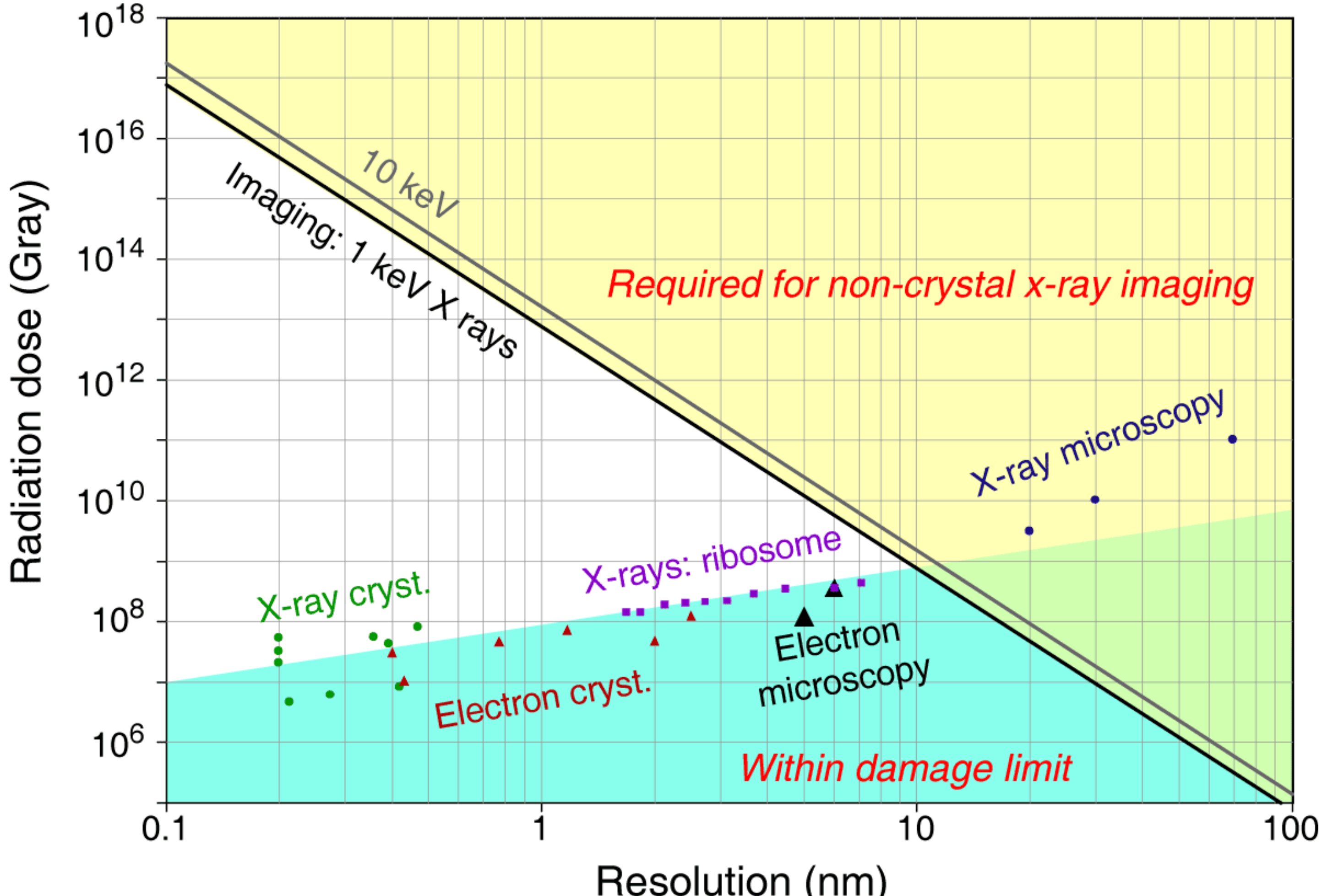
Holographic image of random magnetic domains in a Co/Pt ML sample, Co L_3-edge absorption edge.

S. Eisebitt[1], J. Lüning[2], W. F. Schlotter[2,3], M. Lörgen[1], O. Hellwig[1,4], W. Eberhardt[1] & J. Stöhr[2] NATURE, 432, 885 (2004)

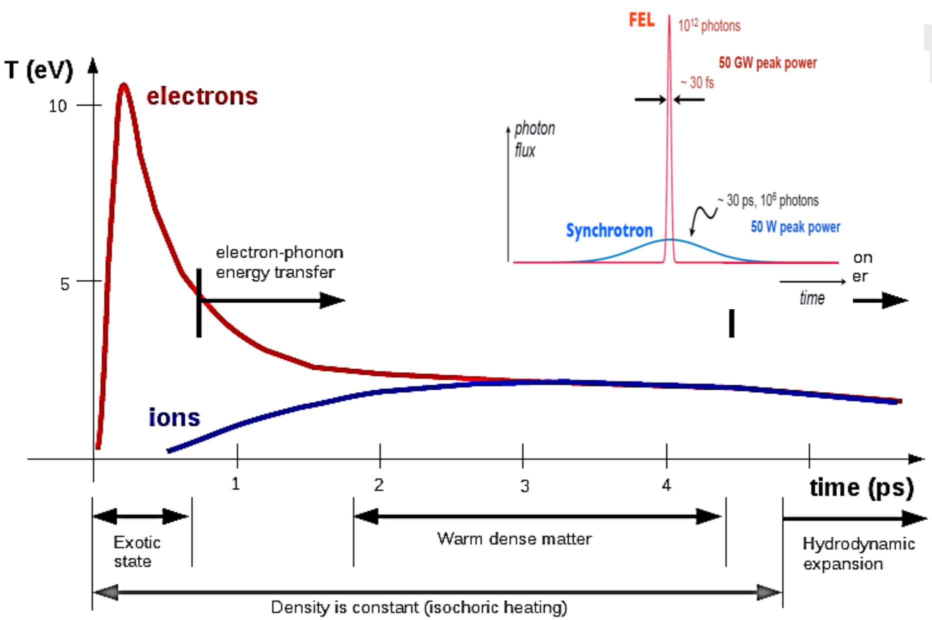


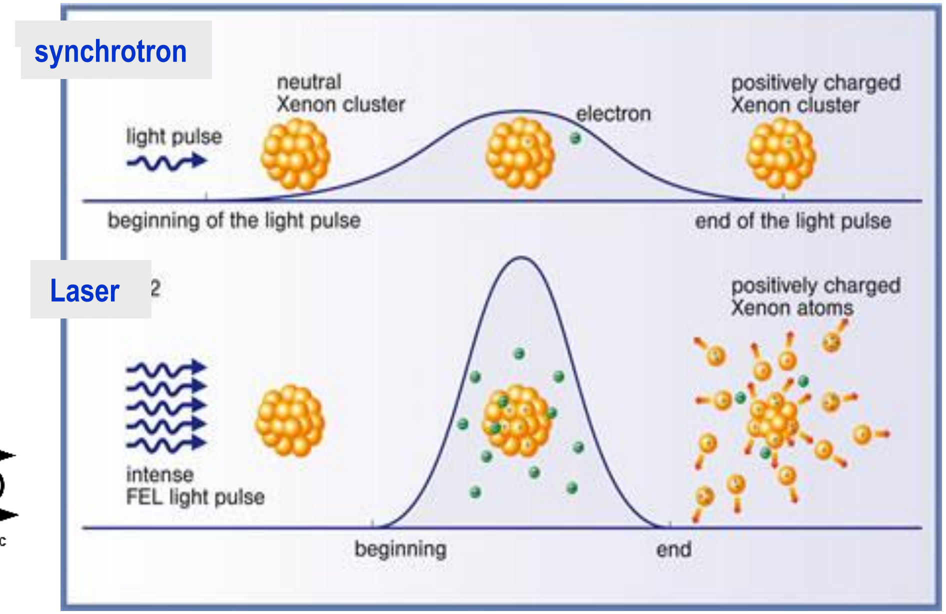
X-ray exposure determines the achievable resolution. Radiation damage sets the dose

For each scattered photon that contributes to the diffraction pattern there are about 10 x-ray photons absorbed. This absorption deposits energy into the sample and leads to sample degradation.



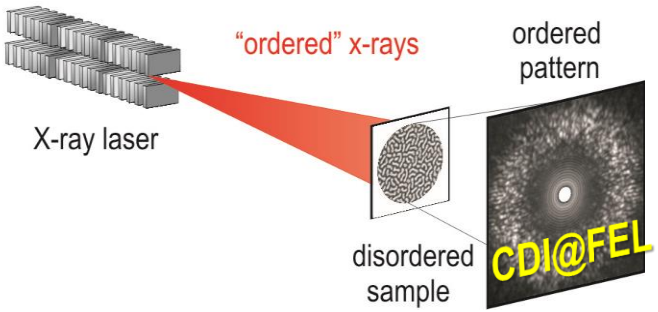
How matter will respond when exposed to a very high power short fs pulses





When matter is irradiated with very intense light, exciting deeper electronic levels, unusual processes occur which do not happen upon irradiation with less intense light: exotic non-equiliblium state with electrons at tens eV temperatures and ions at RT (< 100 fs), electron-phonon energy transfer leading to warm dense matter (>1 ps), lattice expansion (> 5 ps) and Coloumb explosion..

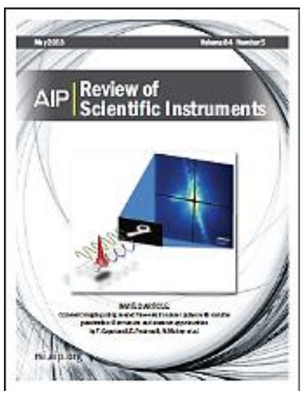
Single shot Coherent Diffraction Imaging (CDI)

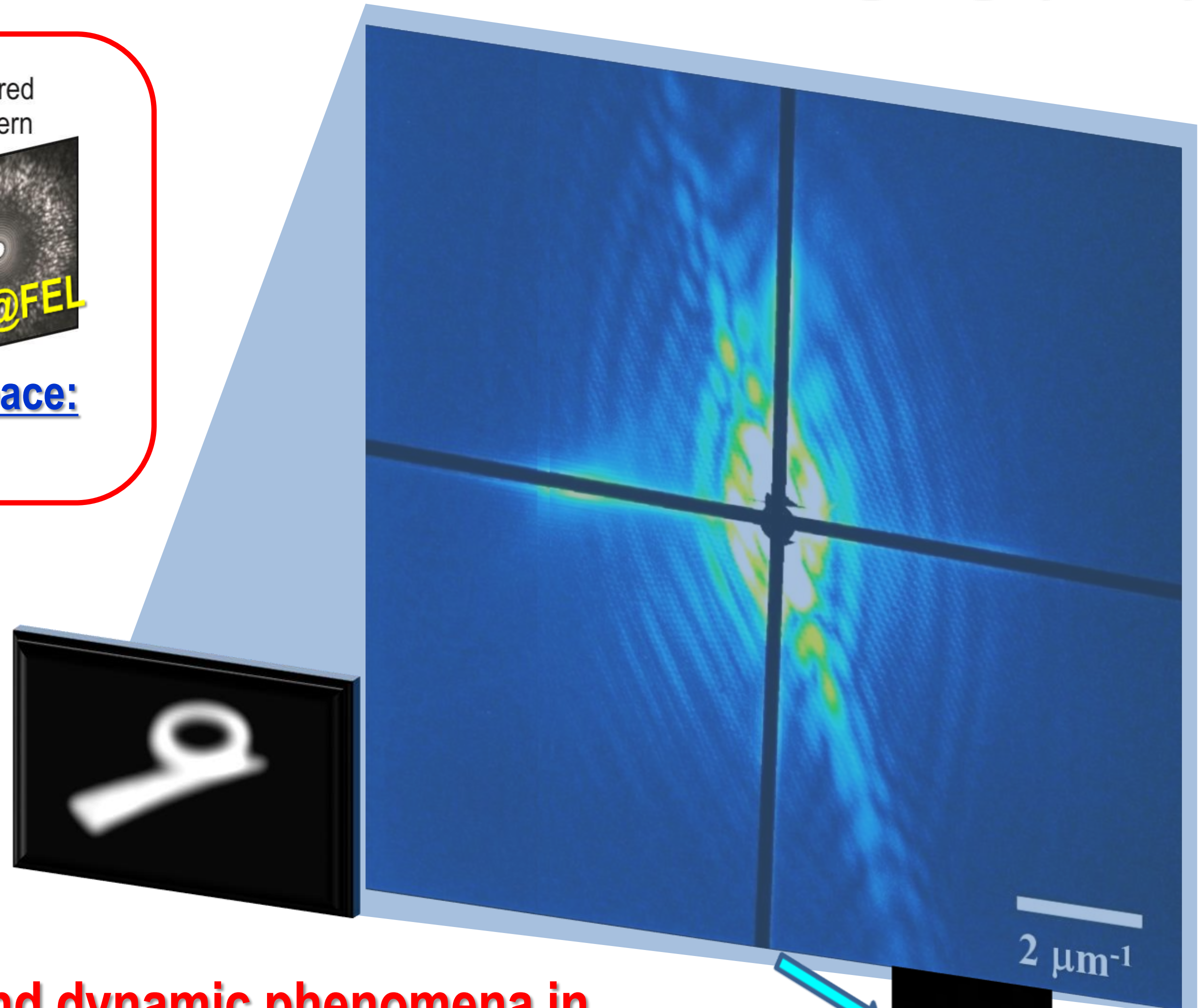


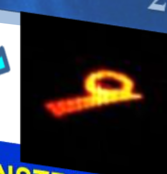
Acquire data in reciprocal space:

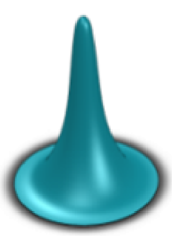
Resolution: δ=λ/sinθ

Chapman et al, (2006)





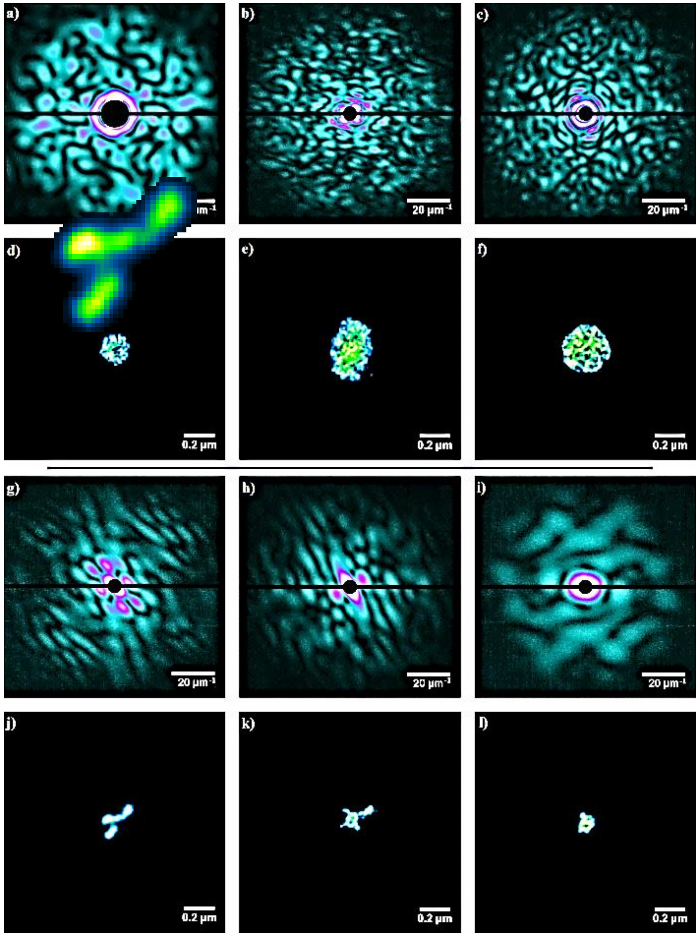




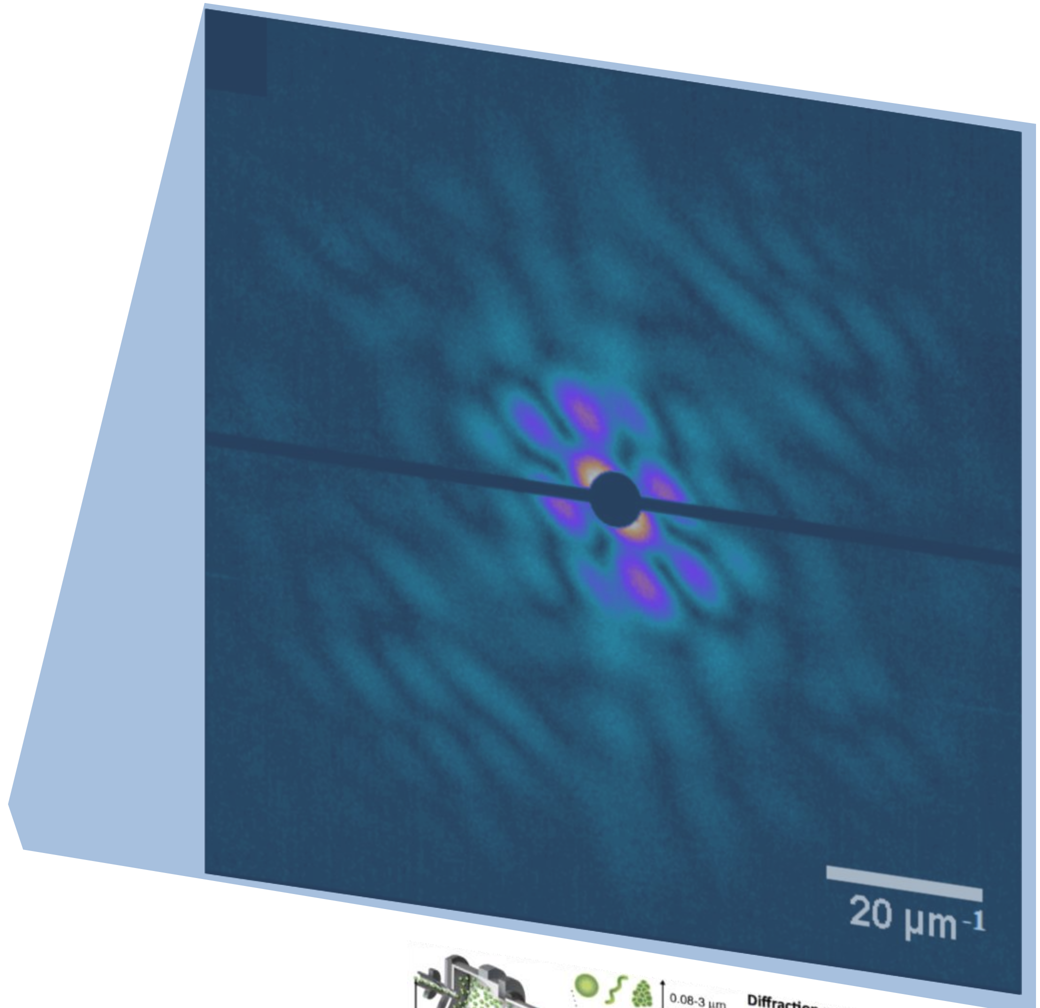
RECONSTRUCTED OBJECT using phase retrieval

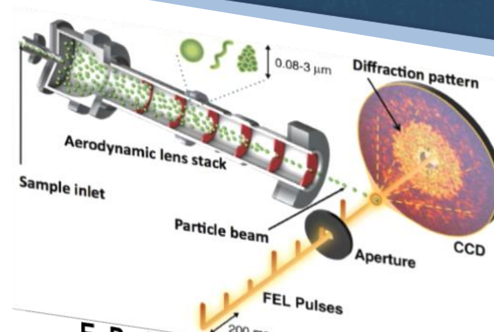
Structure and dynamic phenomena in morphologically complex, disordered or particulate matter – Fermi's spectral purity is an asset!

Elettra Sincrotrone Trieste



Appealing to explore the new collective properties resulting from the secondary structures of the assembled NP





E. Pedersoli et al,
J. Phys. B: At. Mol. Opt. Phys. 46 (2013) 164033

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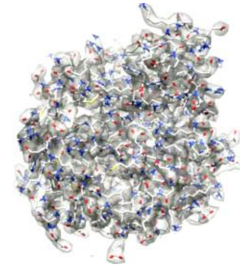
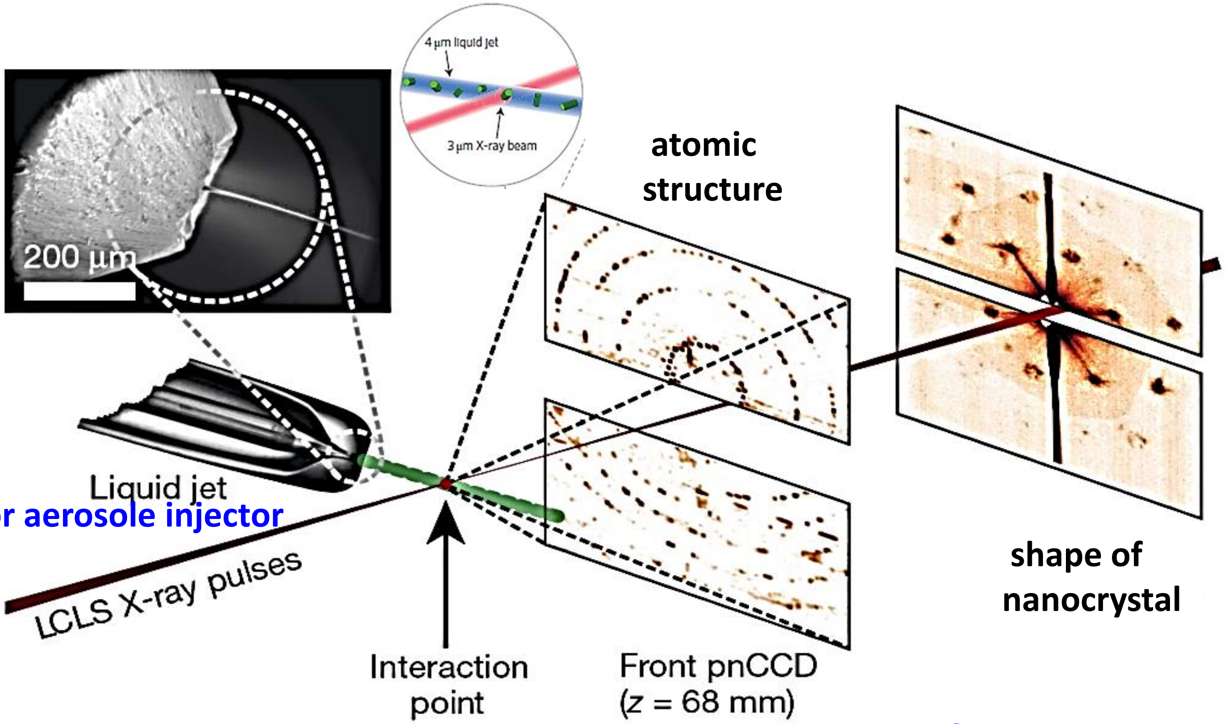
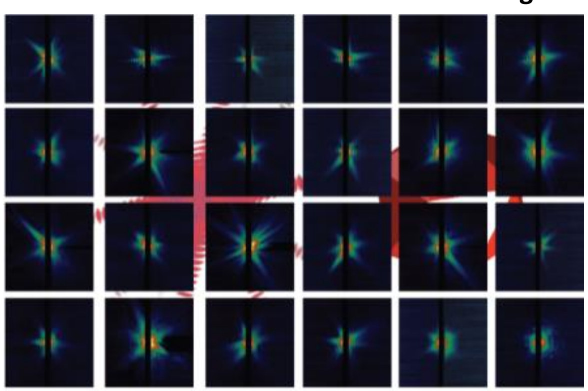
Fs Serial Protein Nano-crystallography

NEXT single molecule CDI

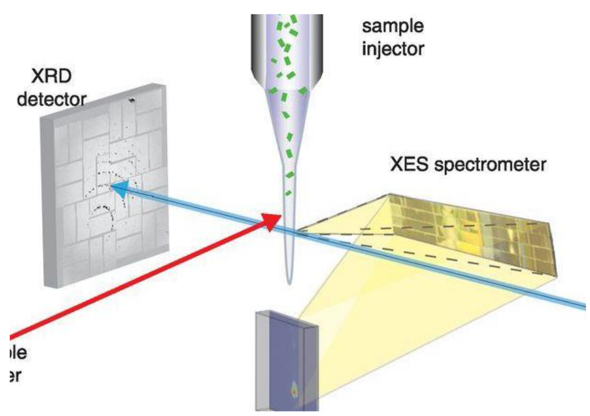




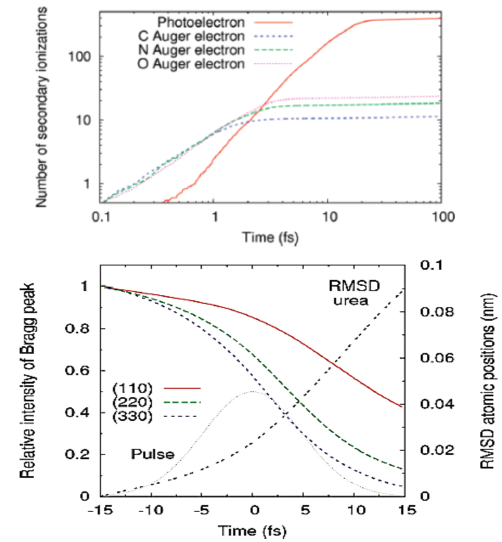
Randomly oriented patterns to be assembled to recover 3D image



TR + Spectroscopy



Need very short pulses

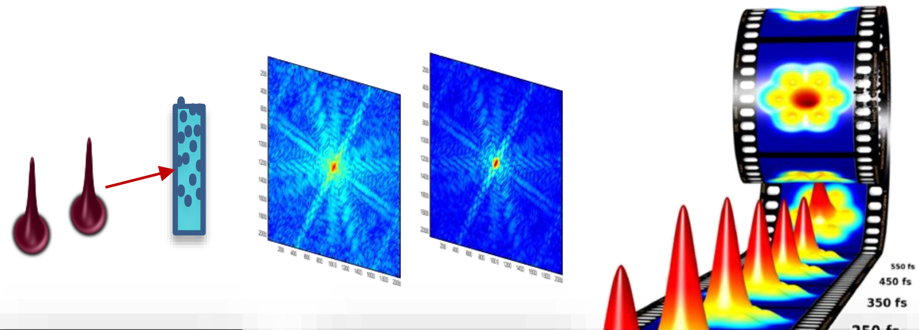


FEL=~ 6-8 keV: radius of gyration of the photoelectron and Auger clouds can reach 300 nm and 8 nm: photoelectron cascade becomes bigger than nano-objects.

C. Caleman et al, ACS NANO 5, 136, 2011

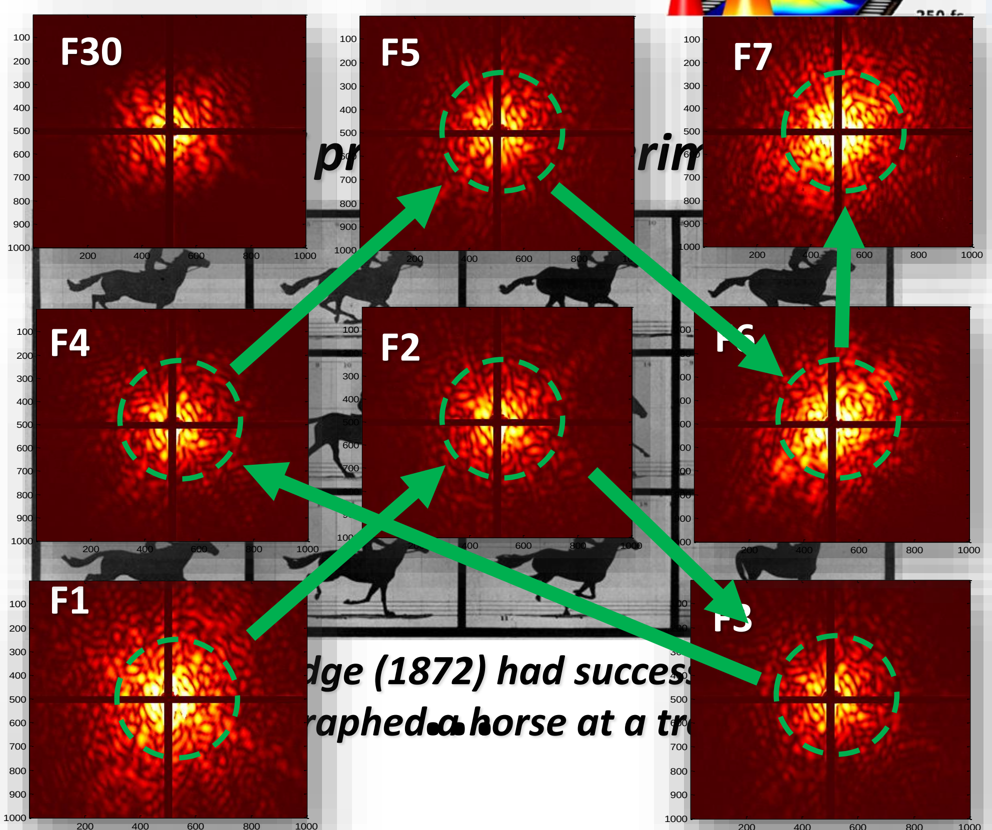
Elettra Sincrotrone Trieste

Towards *fs*-movie with single shot CDI

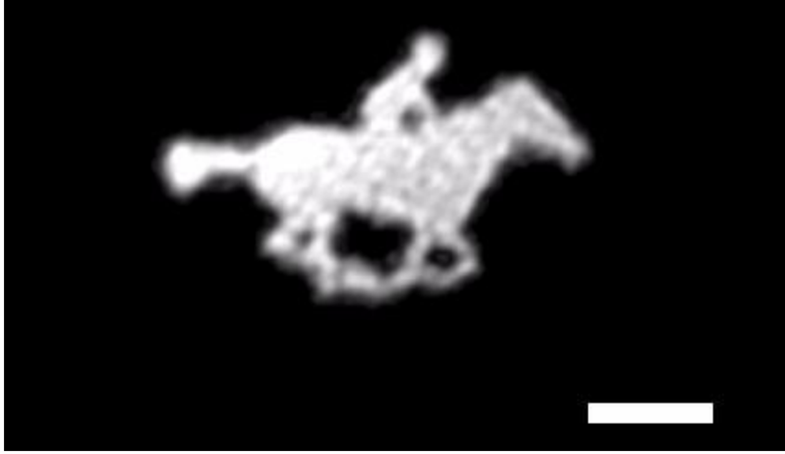


Challenging computational task:

- **Collection of a speckle pattern not a real space image. Phase retrieval algorithm.**
- **Reconstruction algorithm has to catalog orientation and "recognize" the frames temporal evolution.**



Using phase retrieval algorithm: galopping horse movie can be reconstructed



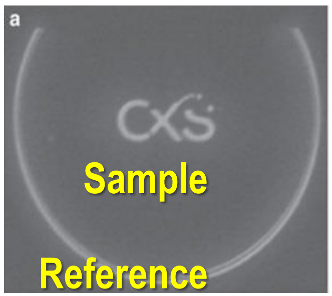
C.H.Yoon et. al. Optics. Exp. 22, 8085 (2014)

Longitudinal coherence of FEL pulses paves a new road to extended reference holography

Elettra Sincrotrone Trieste

FTH ideal case is to optimize the experimental geometry without restriction due to the reference wave, permitting to optimize signal-to-noise and resolution.

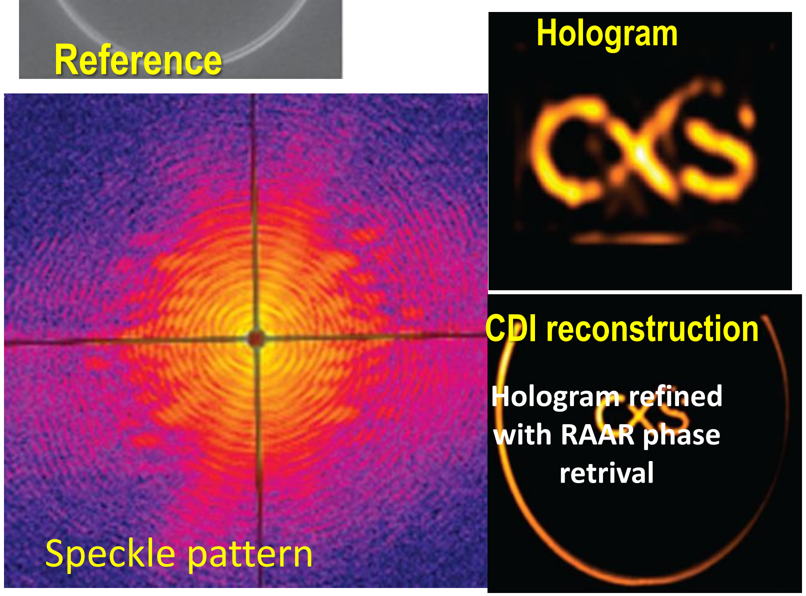
Fourier holography with 'unrestricted choice' of reference wave.



Hologram obtained solving iteratively

$A_{data} - A_{ref} = C_{ref,obj}$

where $C_{ref,obj} = \Psi_{ref}^* \, \Psi_{obj}$

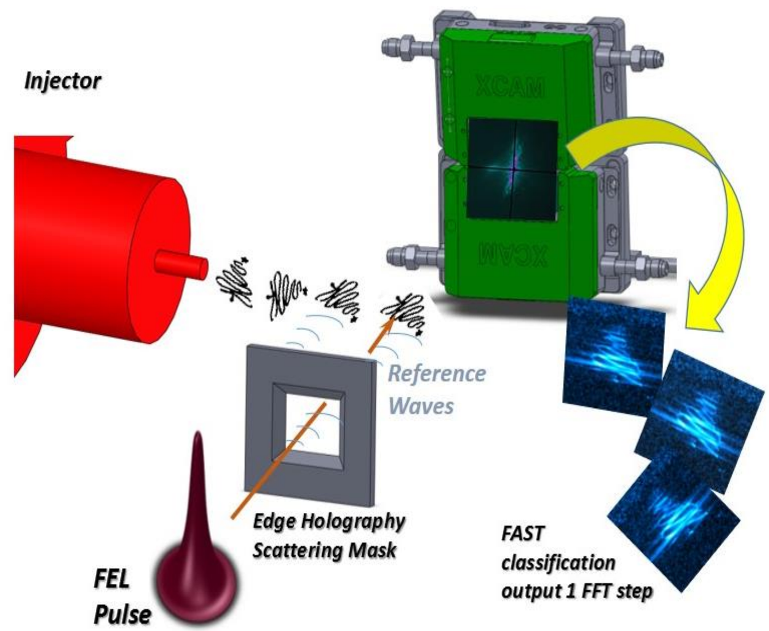


A.V. Martin. et al. Nat. Comm. 4, 2476 (2014)

Seed FEL pulse has high degree of transversal and longitudinal coherence.

1. ***Transversal coherence is the key element in CDI.***
2. ***Longitudinal coherence is important in holography when the mask is decoupled from the sample.***

Combining Extended Reference Holography with particle injector



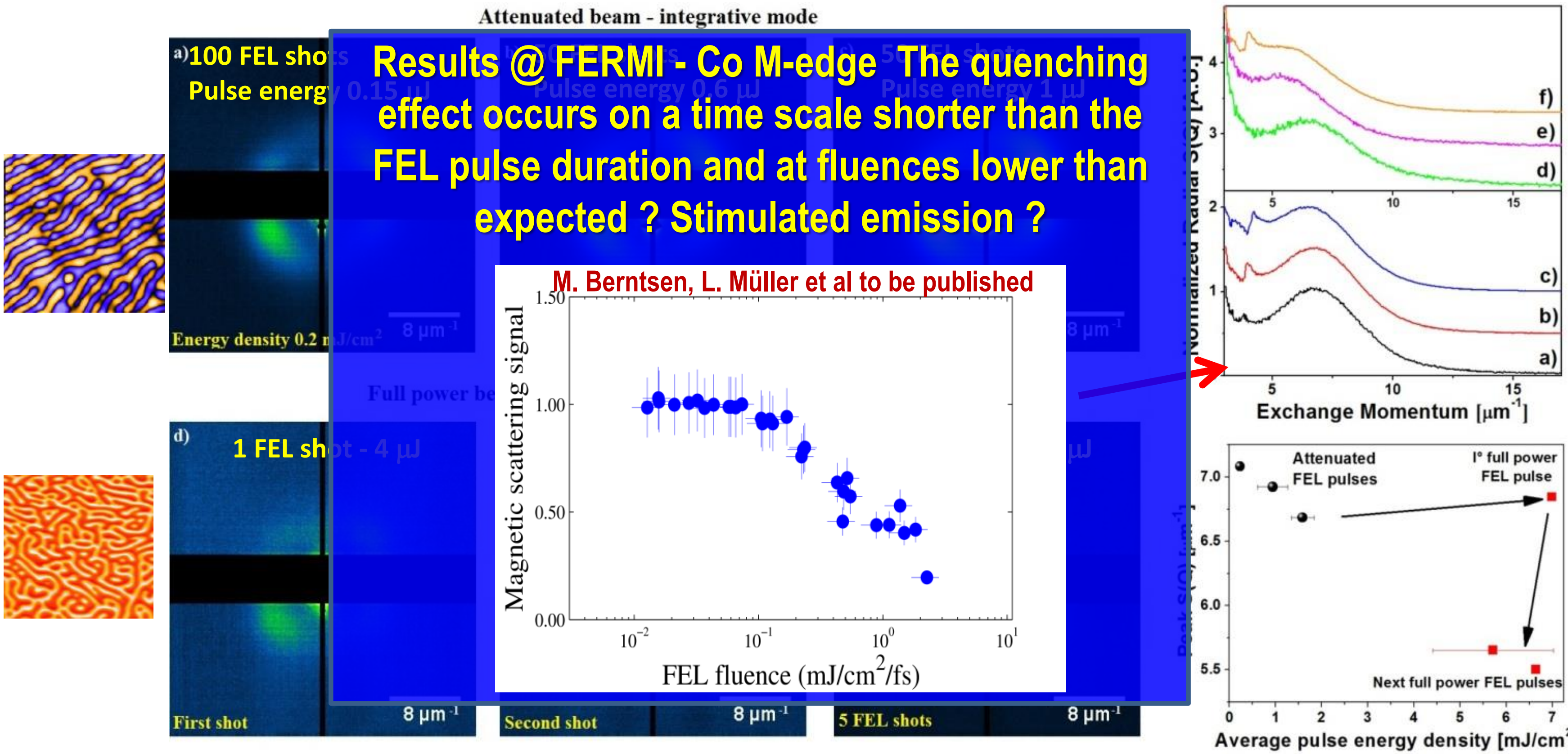
F. Capotondi et al. Optic Express 20 (2012)

‘Static experiments’ affected by the ultrafast events within pulse duration

Elettra Sincrotrone Trieste

FEL radiation not only acts as a probe but also strongly interacts with the sample ? is ‘damage’ threshold for dynamic magnetic studies, oxide reduction…

Reorganization of magnetic domains from aligned to labyrinth structure & change in the average domain period with increasing pulse power:



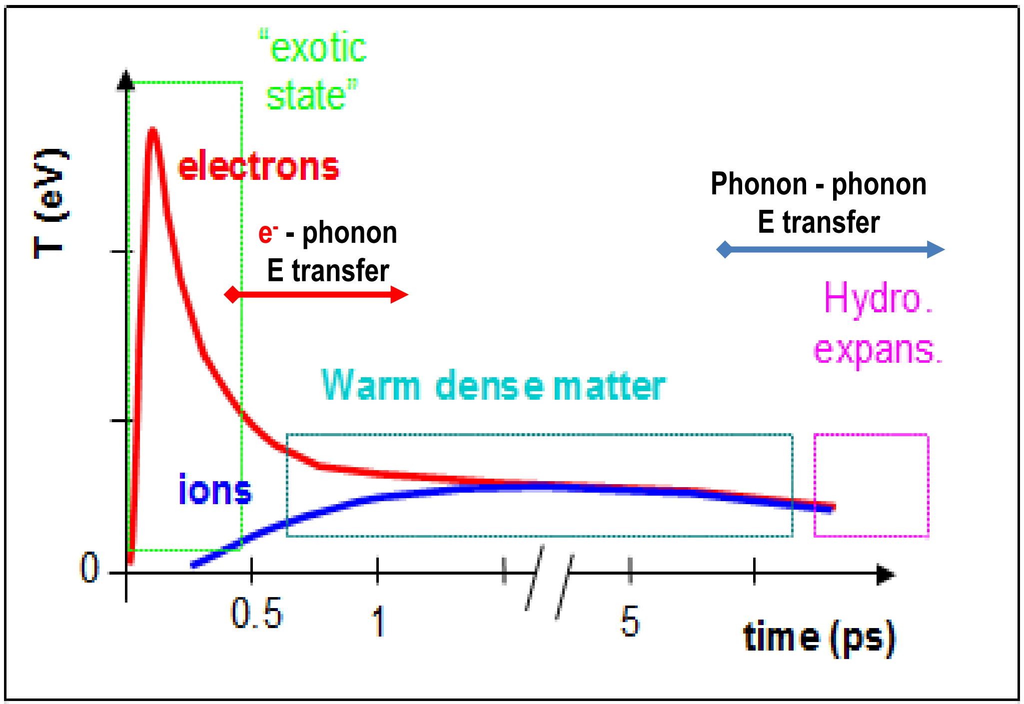
Results @ FERMI - Co M-edge The quenching effect occurs on a time scale shorter than the FEL pulse duration and at fluences lower than expected ? Stimulated emission ?

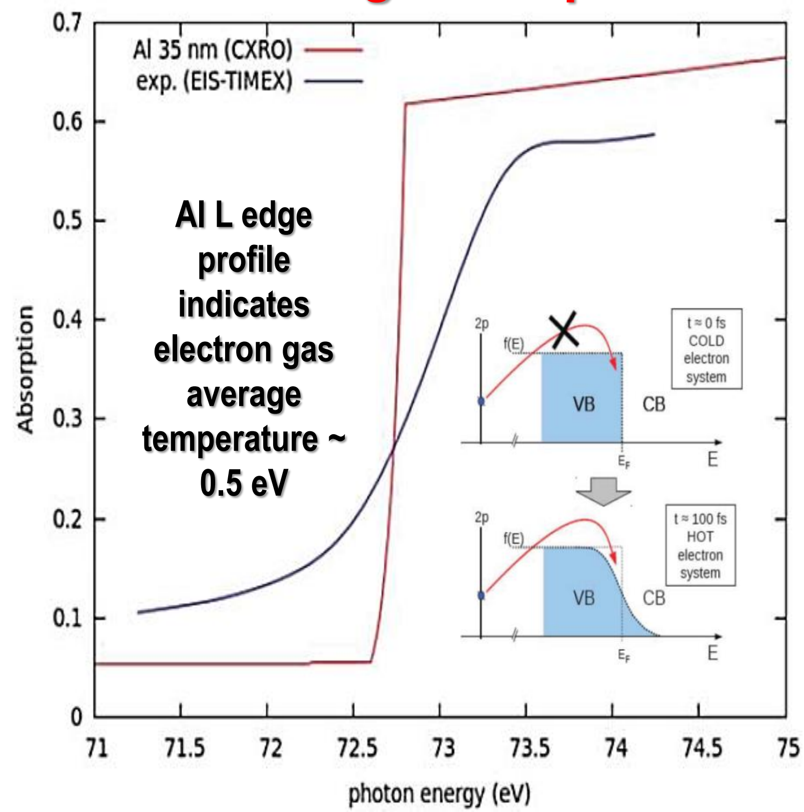
Capotondi et al, RSI 84, 051301 (2013),
Müller et al, PRL, 110, 234801 (2013)

‘Static experiments’: they may be affected by the ultrafast events within the pulse duration

Elettra Sincrotrone Trieste

Ultrafast rearrangement of the electron population across the Fermi level driven by sudden T_e change, encoded in Al L edge XAS profile

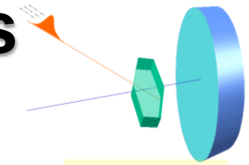




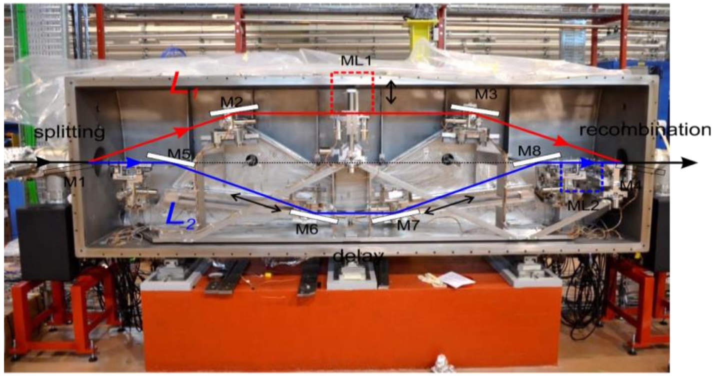
- Thermalization of conduction band electrons occurs within ~ 60 fs pulse duration;
- For E_{photon} above Al 2p edge, the temperature of the electron sub-system is estimated to be ~0.5 eV, well above the Al melting point ~ 0.05 eV.

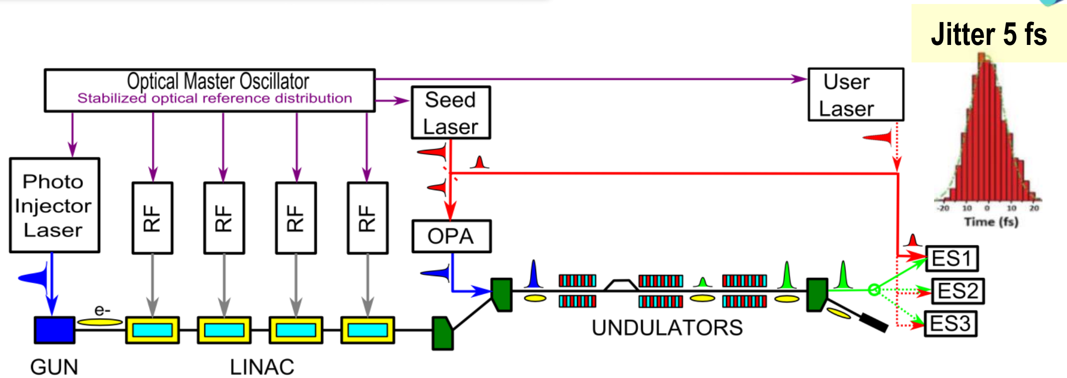
Schemes for stroboscopic experiments

Elettra Sincrotrone Trieste

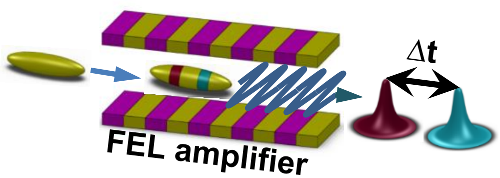


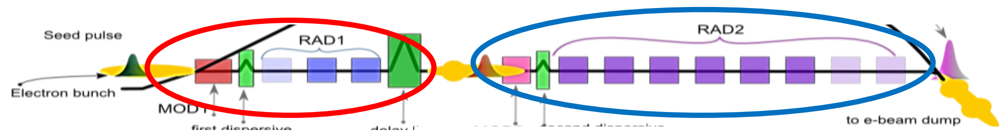
Temporal resolution – fs pulse duration & jitter





P. Cinquegrana et al. Phys. Rev. STAB 17, 040702 (2014) *M. Danailov et al., Opt. Exp. (2014)*





1° stage emits at λ_1 2° stage emits at $\lambda_2 = \lambda_1/N^{arm}$

<u>Single color :</u> FEL-FEL pump/probe pulses with variable intensities using split/delay pulse schemes: <u>-2 ps < Δt < 30 ps</u> or use two electron bunches with a RF cycle temporal distance (Δt ~ 330 ps -10 ns)

<u>Two colour schemes</u>:

- **Optical pump *or probe*** and **FEL *probe* or pump:** "timing jitter issue"- intrinsic synchronization of the seed optical pulse with FERMI pulse is great advantage;
- **Two FEL pulses with different λ_i and variable intensities** via single or twin seed laser schemes (Δt 150-800 fs), or using double undulator modes (Δλ is larger)
- **Two FEL pulses with different λ_i using the first and second stage of FERMI2**

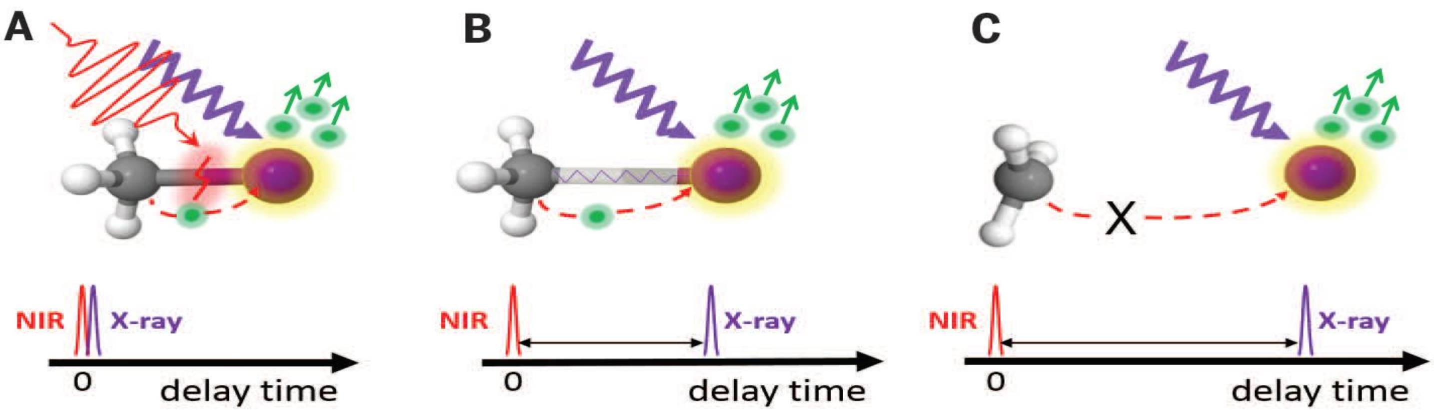
Maya Kiskinova

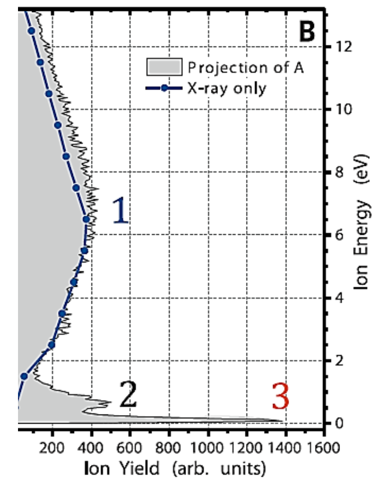
Elettra Sincrotrone Trieste

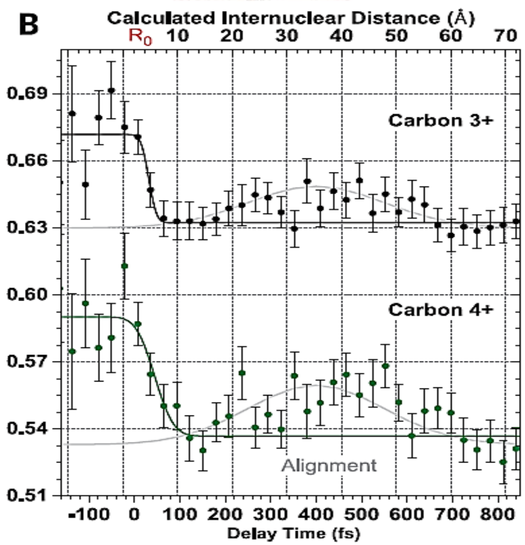
Two color IR pump/FEL probe time resolved FEL experiments:

IR-induced dissociation of CH_3I following the energy of emitted I ions

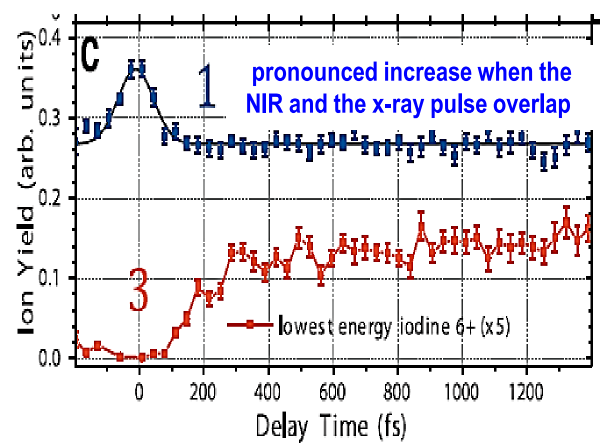
Localized charge on I atom, created by X-rays may transfer to the methyl group via Auger decay: event affected by the I-C distance







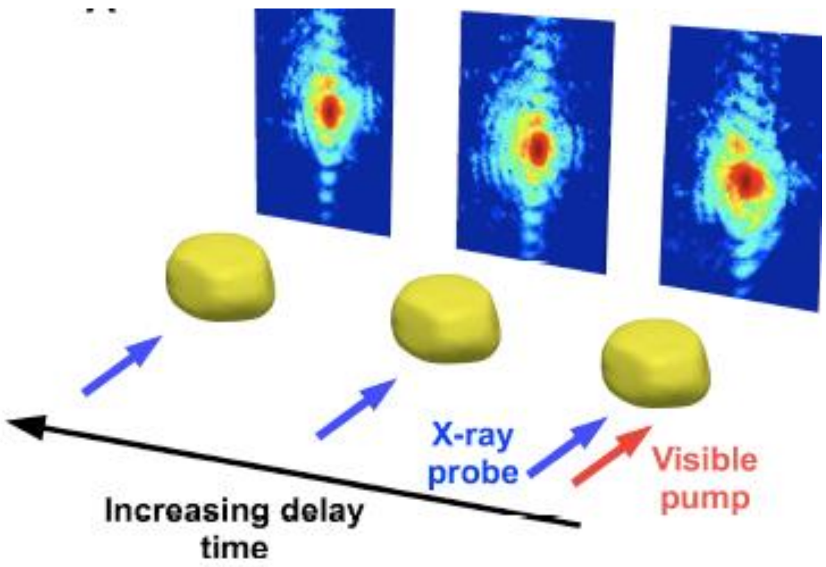
C ions mainly acquire their charge via electron transfer to initially ionized iodine. After ~ 100 fs the molecule charge becomes determined by FEL interaction with CH_3 and I



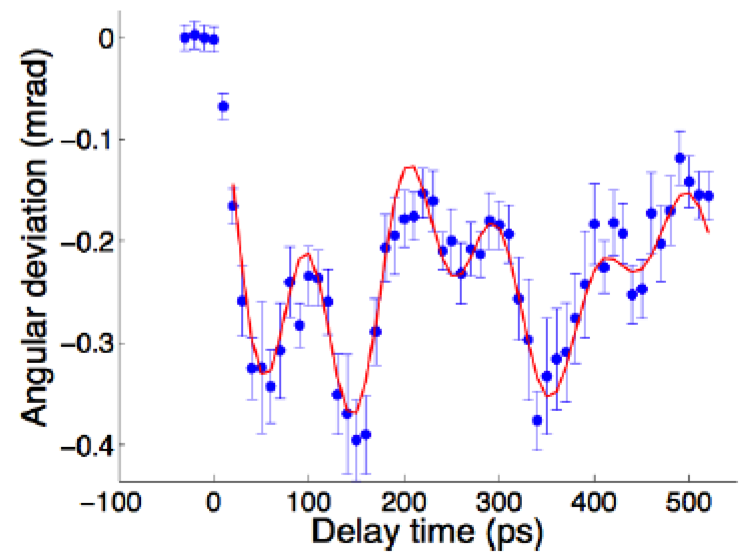
B. Erk, Science (2014)

Two color IR pump/FEL probe time resolved FEL experiments:

Shedding light on lattice dynamics in individual gold nanocrystals via coherent diffraction



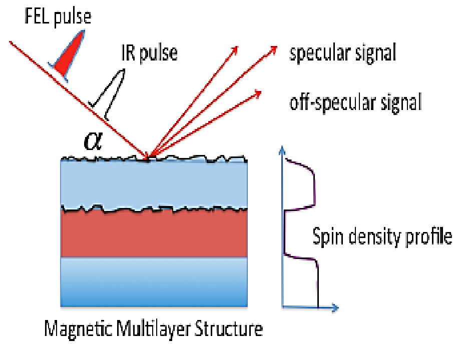
Bragg peak shift with delay time



The evolution of the coherent acoustic phonons within the nanocrystal through the Bragg peak shift: can be modelled as a harmonic oscillator with two modes.

J. Clark, Science 341, 56–59 (2013)

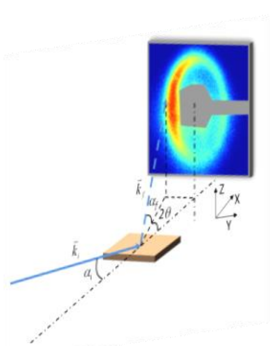
IR pump /FELprobe: time resolved 'magnetic' reflectivity and scattering

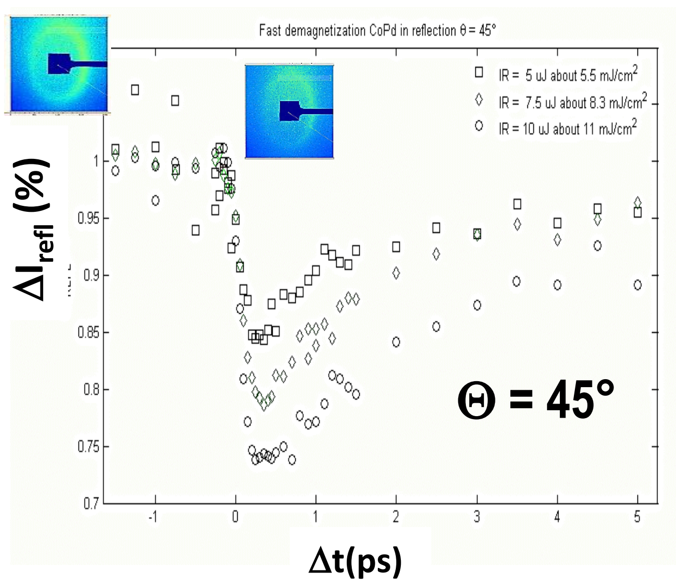


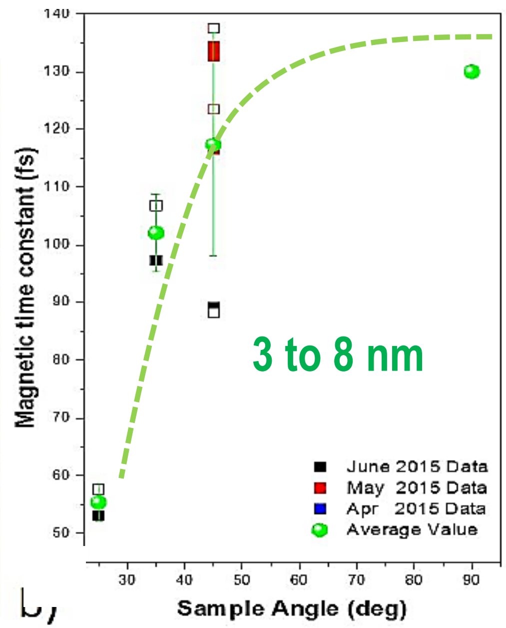
Variable FEL penetration depths as a function of the incidence angle Θ allow for probing after IR excitation:
(1) the effect of spin-diffusion on magnetization profile in vertical direction and (2) the spin transport across interfaces and surfaces.

The scattered photons encode the coexistence of phases, e.g. nucleation of magnetic domains during phase transition.

CoPd ML (Co M-edge): Fast demagnitization as a function of IR fluence



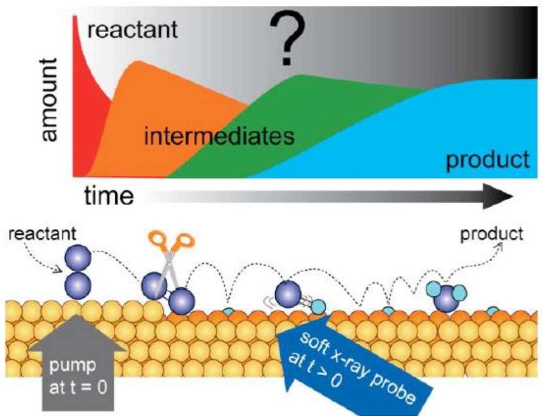


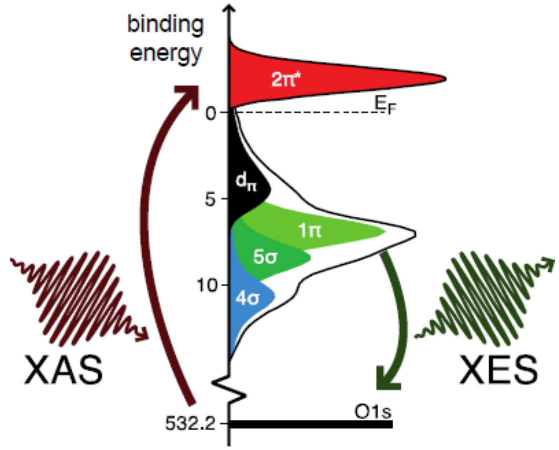


Average value shows a decrease of the demagnetization time when the FEL pulse probes a shallow part of the magnetic structure.

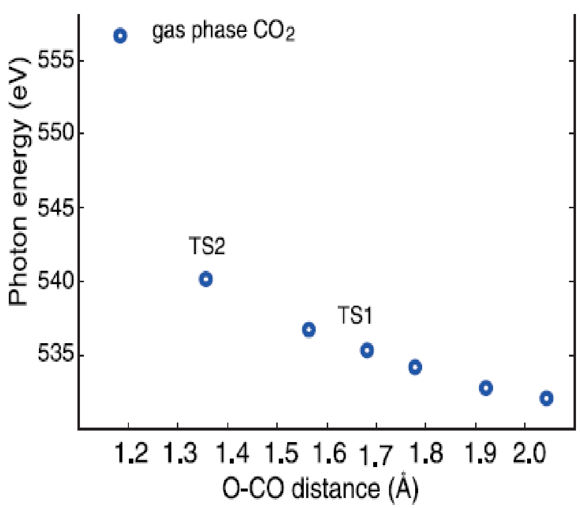
IR pump /FELprobe: Laser-induced surface reactions:

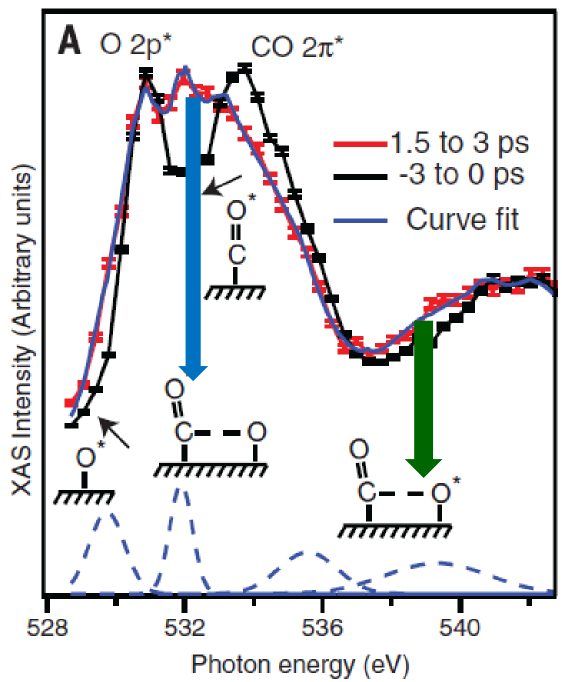
map transient changes of electronic structure by trans (x-ray emission/absorption spectroscopy - XES/XAS)

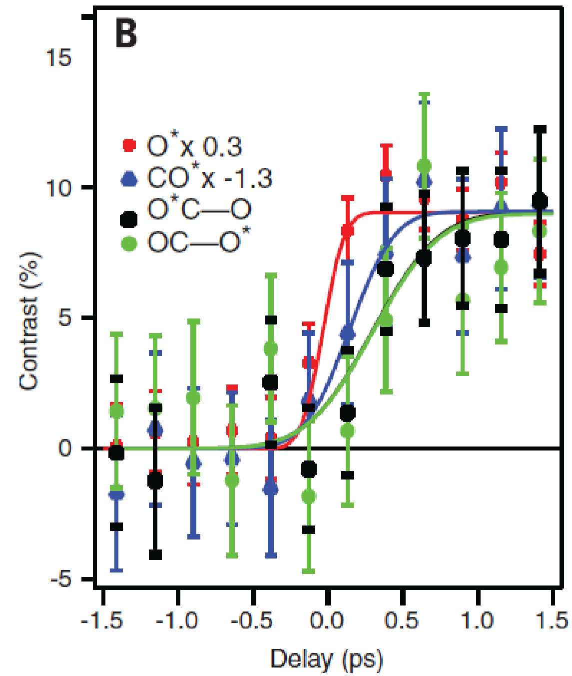




O becomes activated on a time scale below 300 fs, whereas CO is activated on a 500-fs time scale and the transient states 1 and 2 are formed leading to CO_2





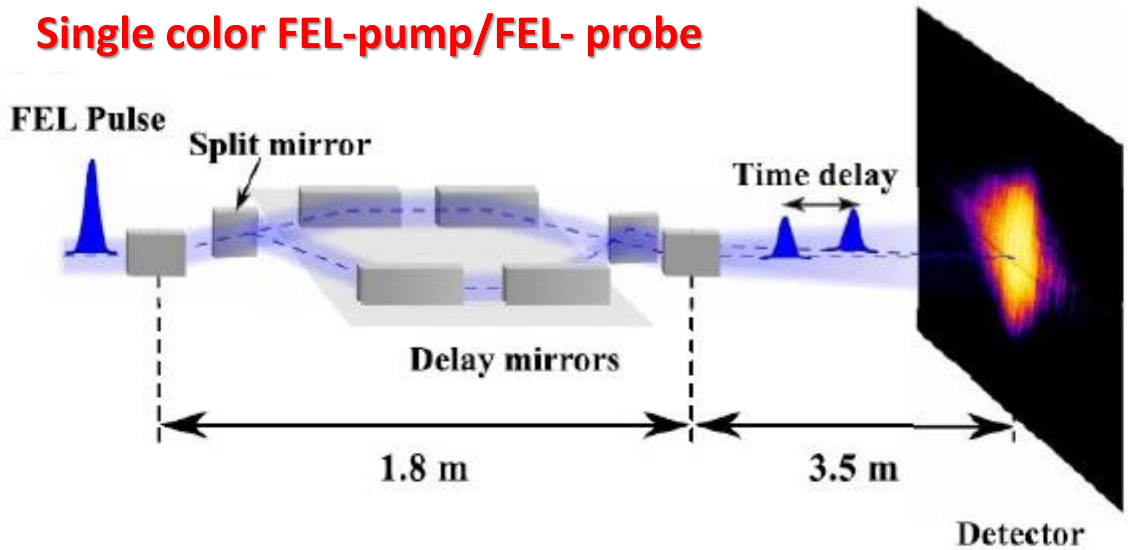


H. Öström et al. Science 347, 978 (2015)

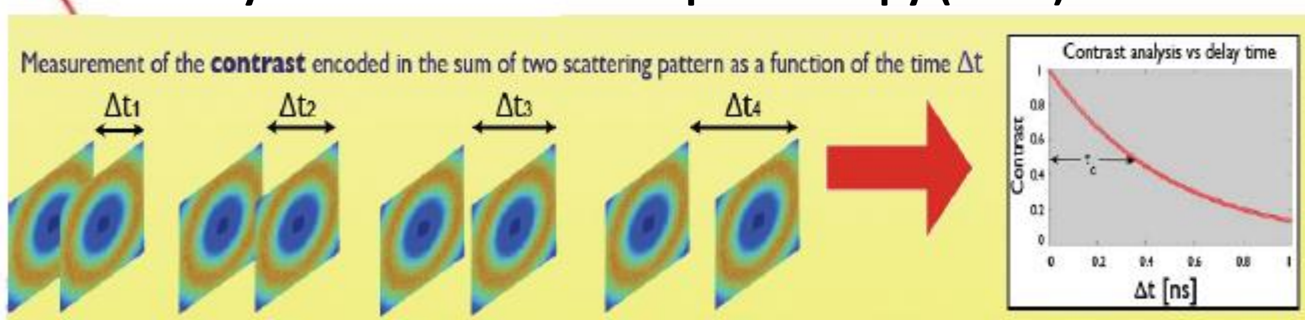
Maya Kiskinova

Single color FEL-pump/probe for schemes for FEL/FEL experiments:

Single color FEL-pump/FEL- probe

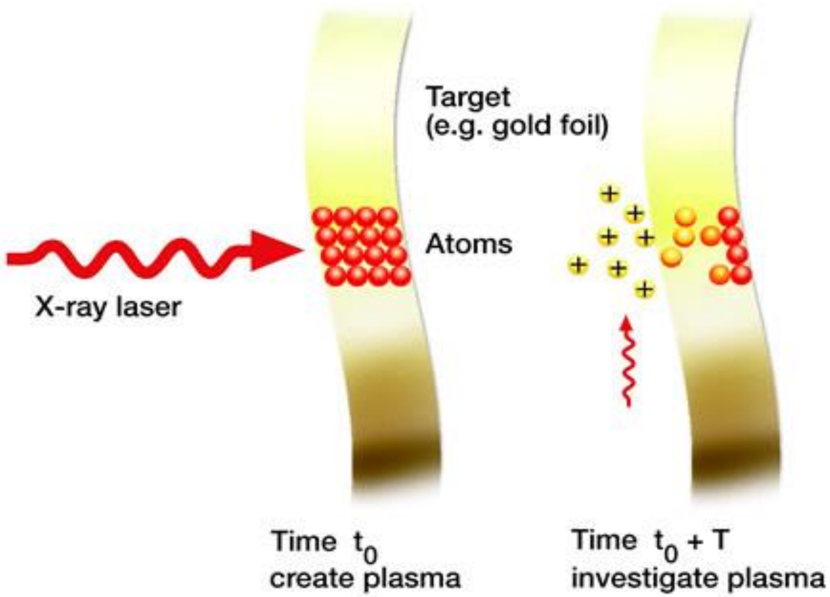


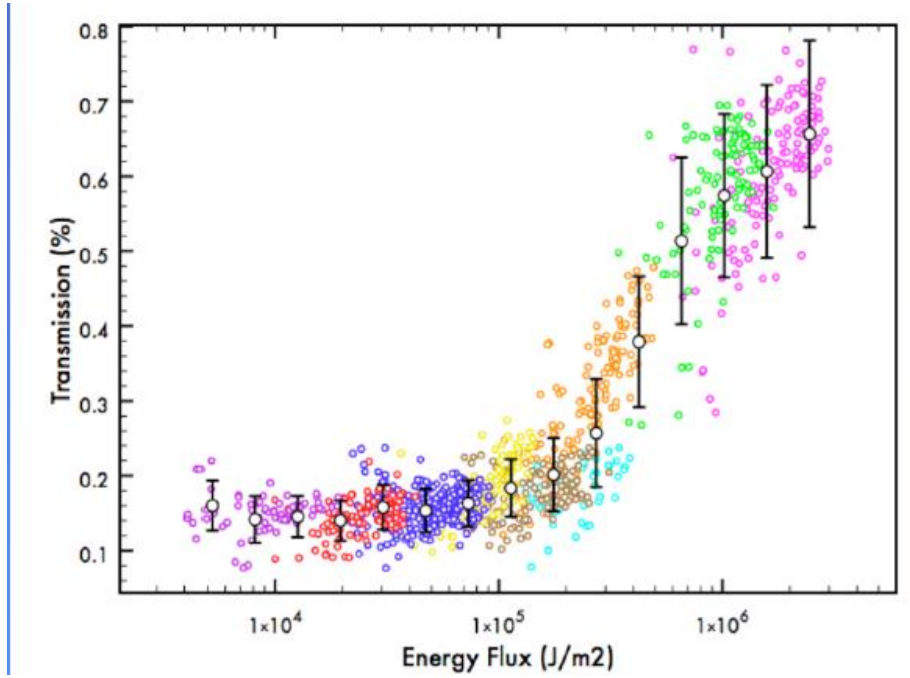
X-ray Photon Correlation Spectroscopy (XPCS)



Producing plasma – highly ionized state of matter and monitoring its evolution via FEL pump-probe

With an X-ray laser, plasmas can be created that are as hot as the interiors of giant stars. At the same time, it will be possible to investigate the status of created plasmas at varying intervals with another part of the laser beam and thus to conduct research into the plasma state.





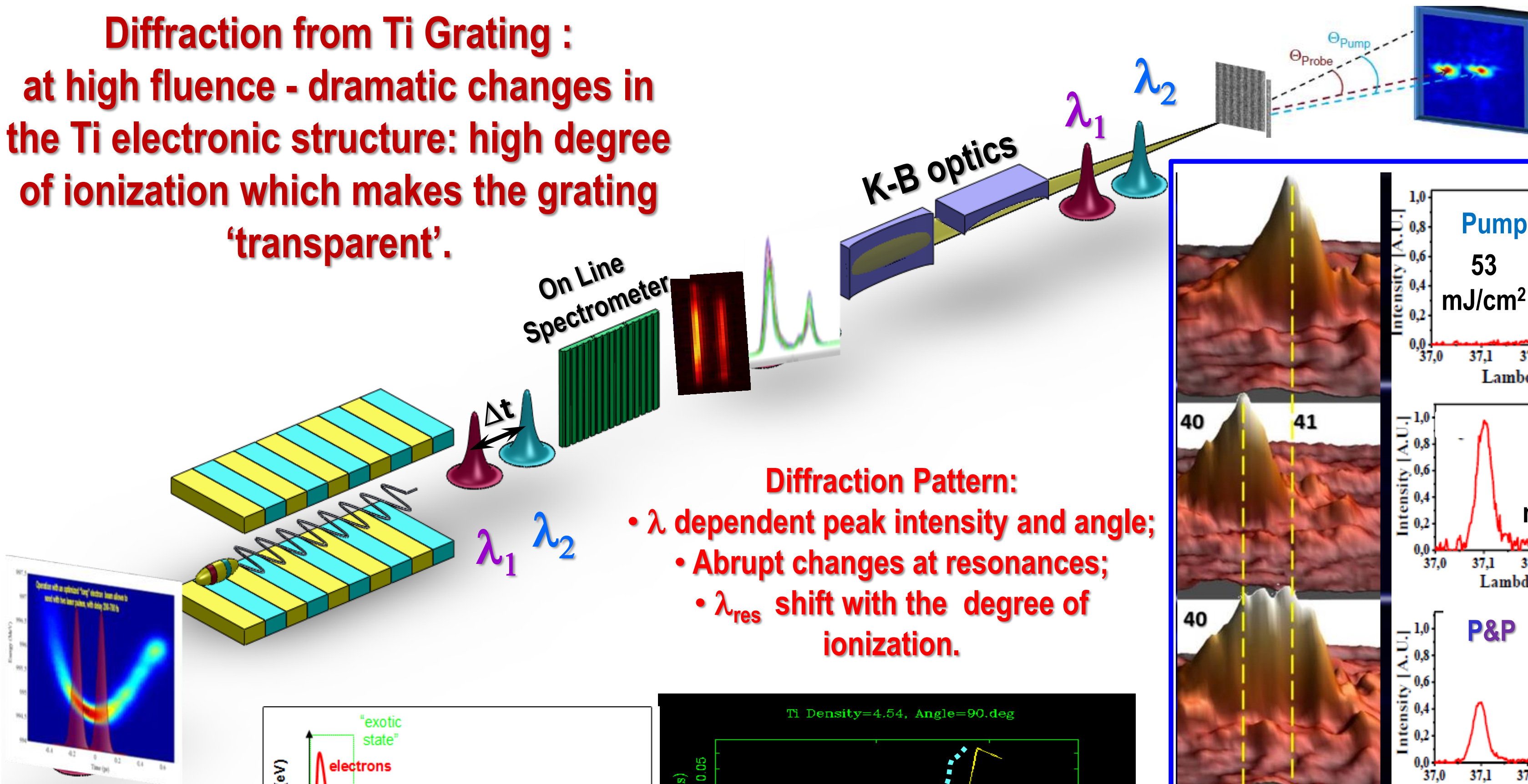
Al foil irradiated with high energy 92 eV FEL photons becomes transparent because both electrons from the 2p state are ejected and no more photoionization of electrons is possible – blue shift in the L edge

Maya Kiskinova

Diffraction from Ti grating using two color FEL pulses

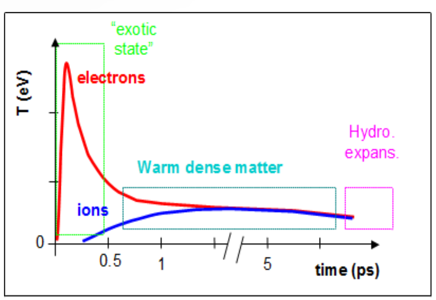
Elettra Sincrotrone Trieste

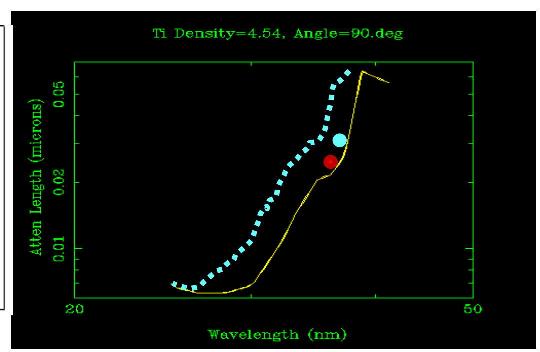
Diffraction from Ti Grating : at high fluence - dramatic changes in the Ti electronic structure: high degree of ionization which makes the grating 'transparent'.

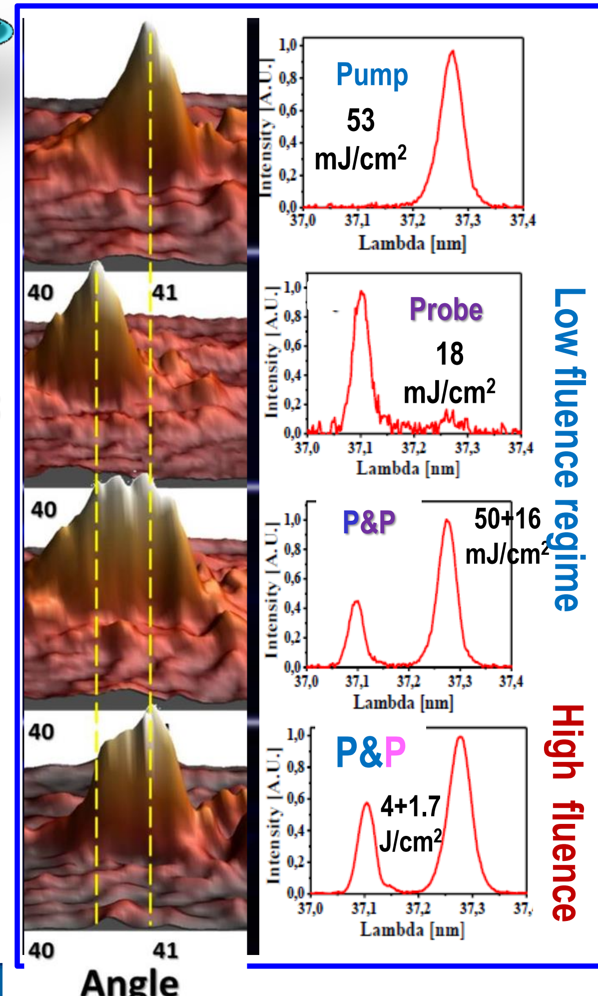


Diffraction Pattern:

- λ dependent peak intensity and angle;
- Abrupt changes at resonances;
- λ_{res} shift with the degree of ionization.





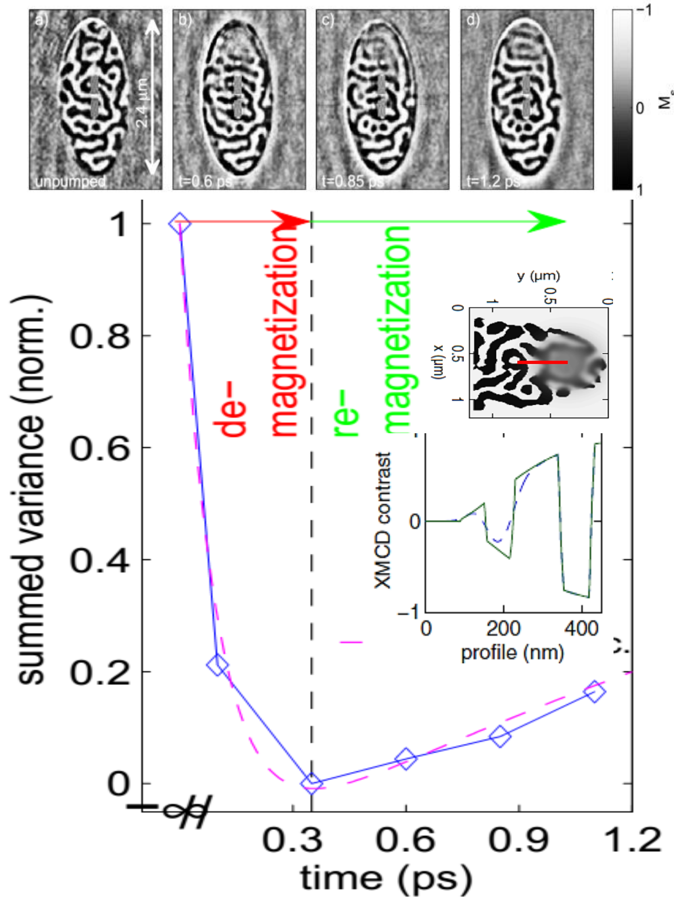


NATURE COMMUNICATIONS | 4:2476 | DOI: 10.1038/ncomms3476 |

Maya Kiskinova

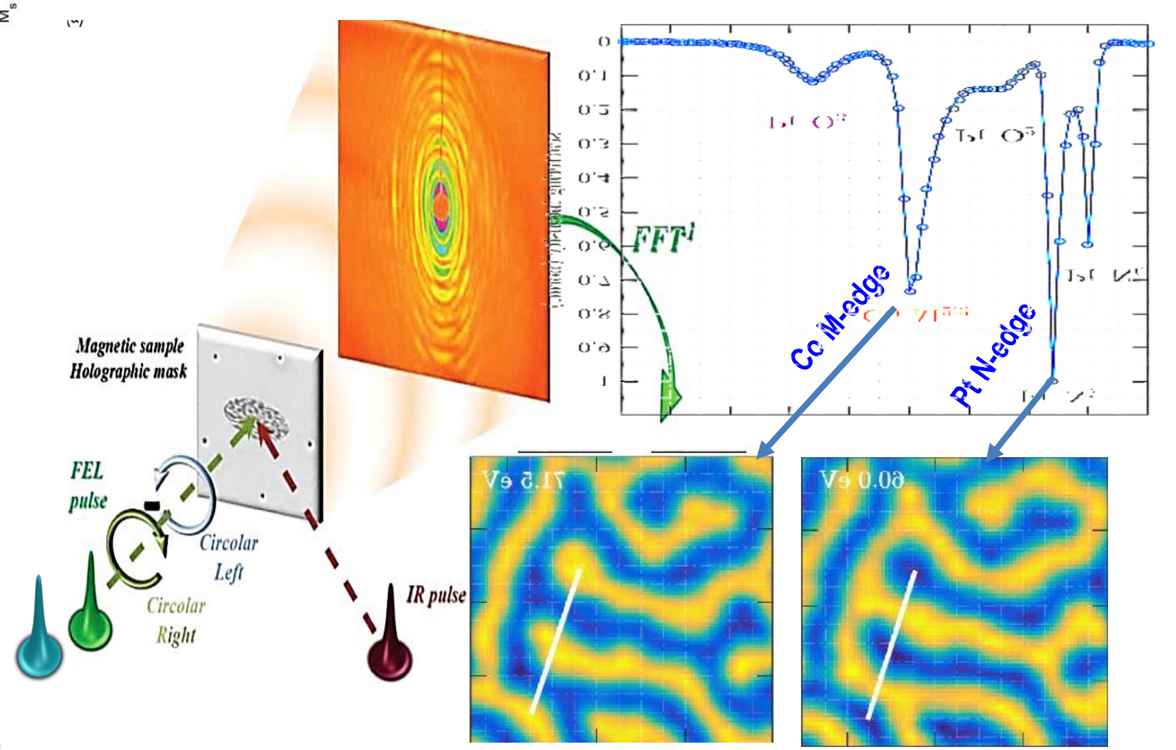
Elettra Sincrotrone Trieste

IR pump /FELprobe: Time resolved resonant magnetic holography with sub-100 nm spatial and 100-fs temporal resolution difference



Demagnitization dynamics after IR pump

FTH: Two color element-specific magnetization in Co/Pt heterostructures



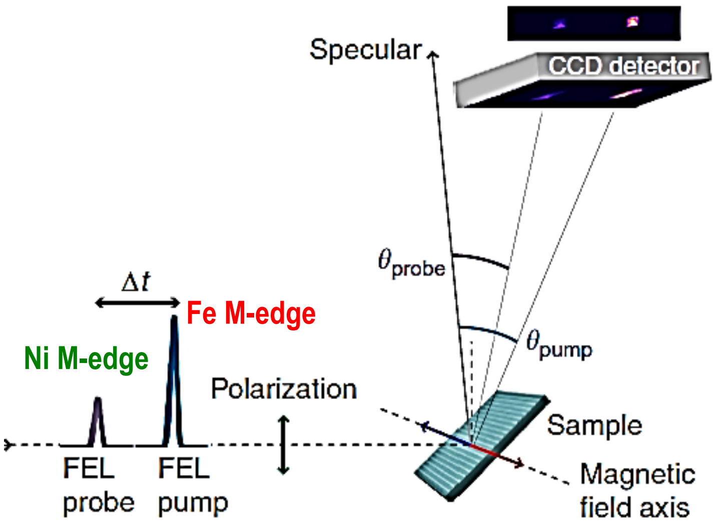
Pt and Co layers magnetic domain patterns reconstructed from a single difference hologram: "inverted domains" – induced magnetization in Pt layer.

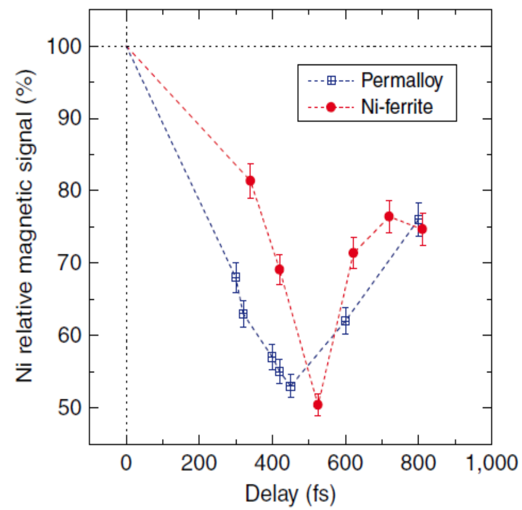
C. von Korff Schmising et. al. Phys. Rew. Lett. (2014).;
Structural Dynamics 4, 014301 (2017);

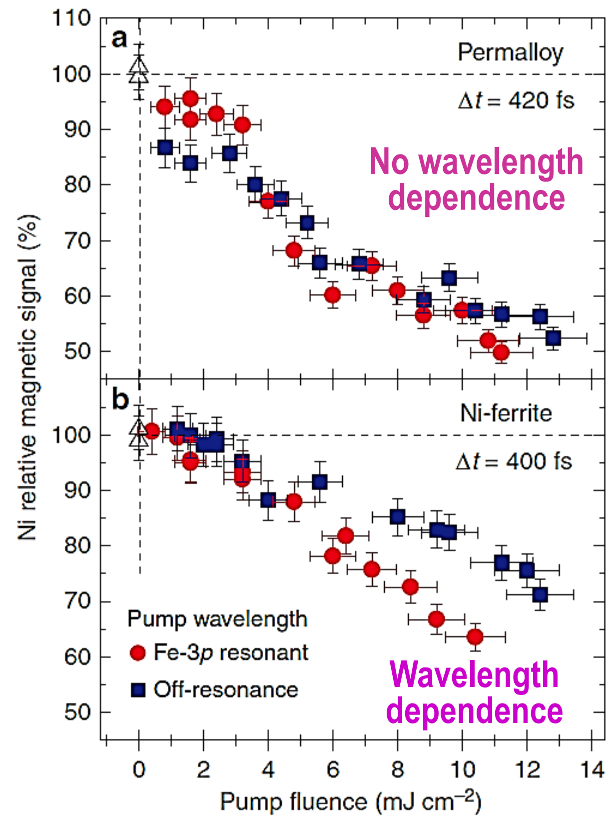
Elettra Sincrotrone Trieste

Two-colour FEL: double resonance experiment with NiFe and $NiFe_2O_4$ magnetic grating samples

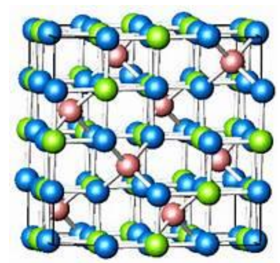
nature COMMUNICATIONS *E. Ferrari et al., (2016)*







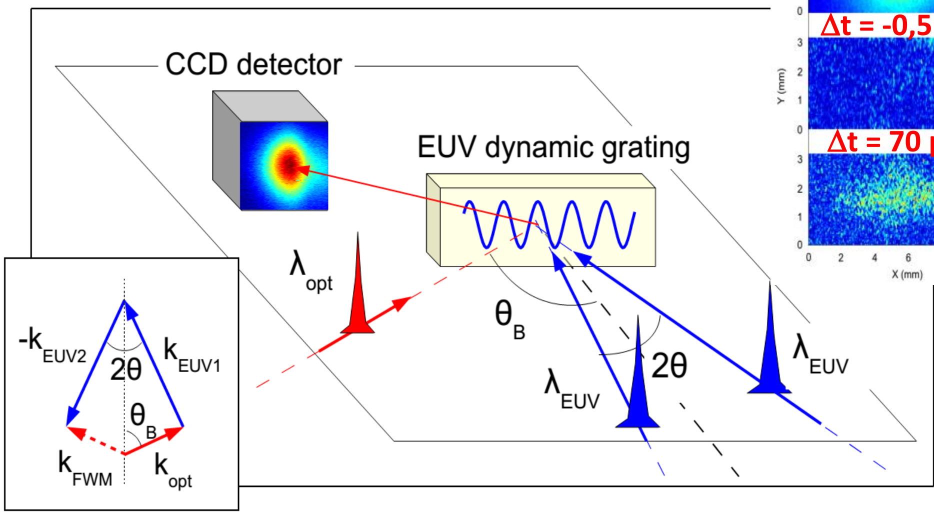
Fe and Ni 3d orbitals: delocalized in permaloy localized in Ferrite (O-mediated exchange expected)

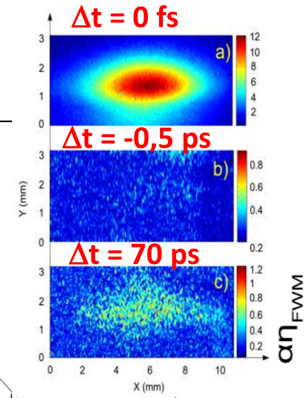


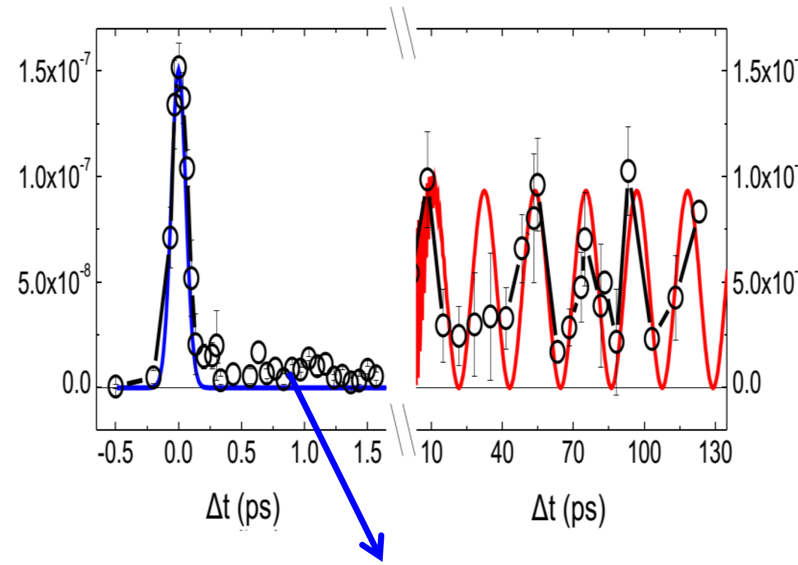
Magnetization dynamics in oxides and other complex magnetic materials with highly localized orbitals and mediated coupling can be sensibly affected by tuning the pump energy to a specific electronic excitation of selected atomic edges…

Four wave mixing: generation of EUV transient grating using two coincident FEL pulses

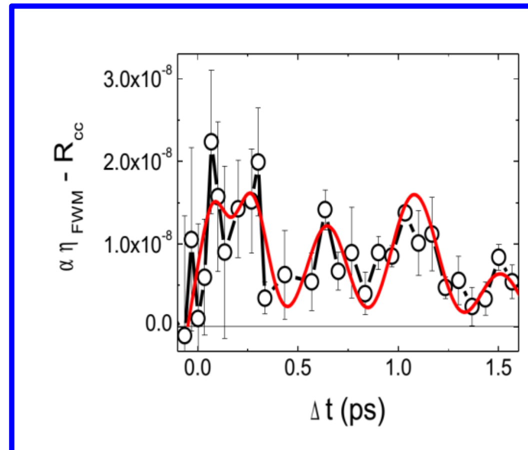
Elettra Sincrotrone Trieste





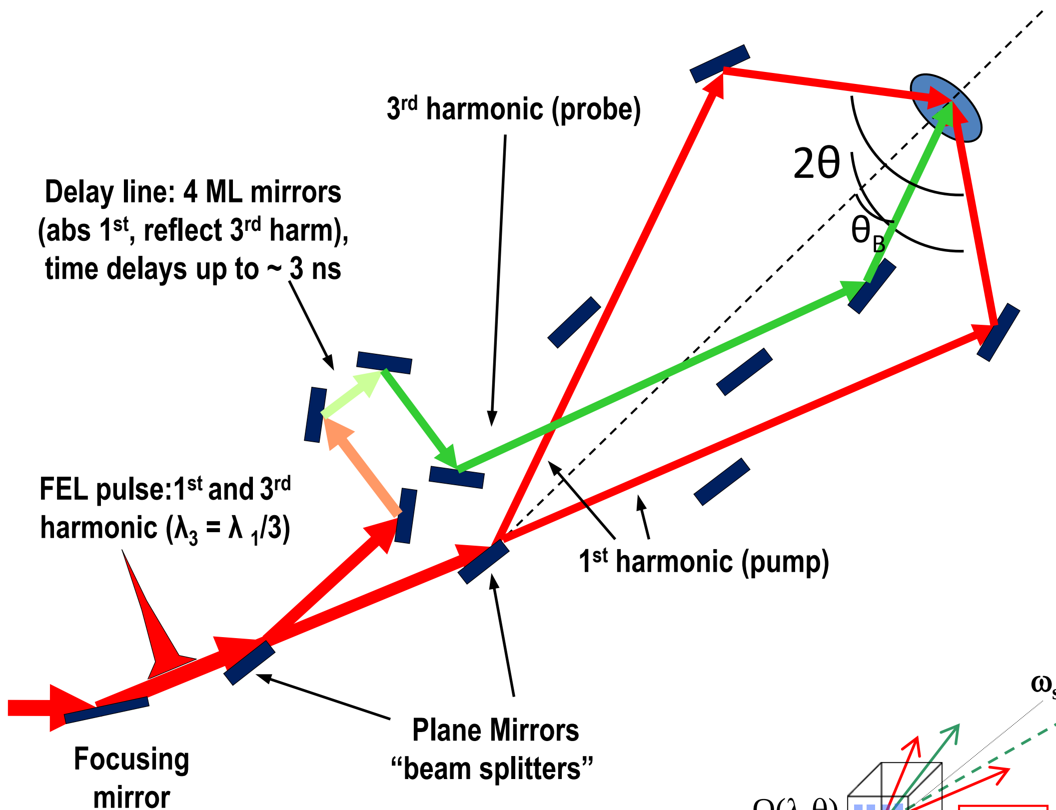


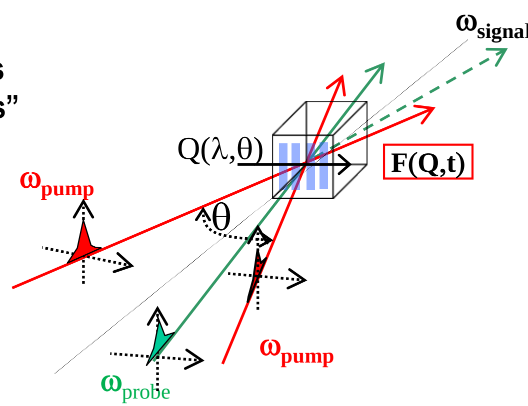
When the three beams arrive on the sample at the same time (Δt = 0) a FWM signal is recorded, showing the occurence of the wave mixing process. With time delay of the optical pulse, intensity modulation of the scattered signal from SiO_2 sample are observed compatible with the excitation of Raman modes (Δt < 1.5 ps) and longitudinal acoustic modes (Δt > 10 ps).



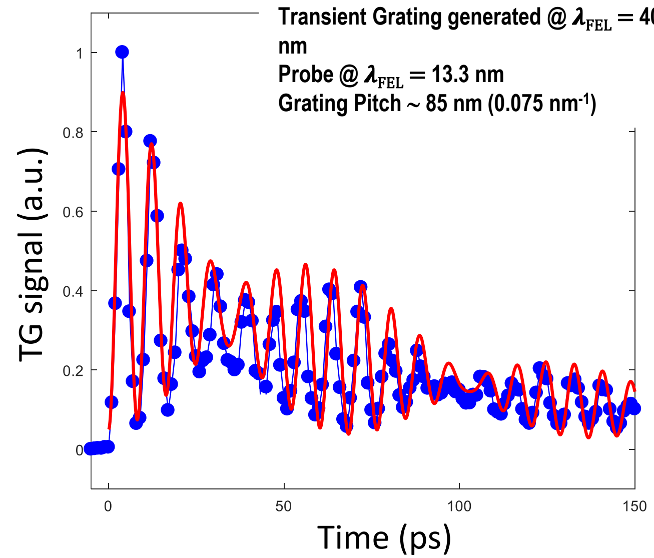
Maya Kiskinova

TIMER@FERMI: Unique beamline for coherent transient grating experiments





Acustic phonon oscillation in Si_3N_4 thin film probed in X-TG- Sept2017!

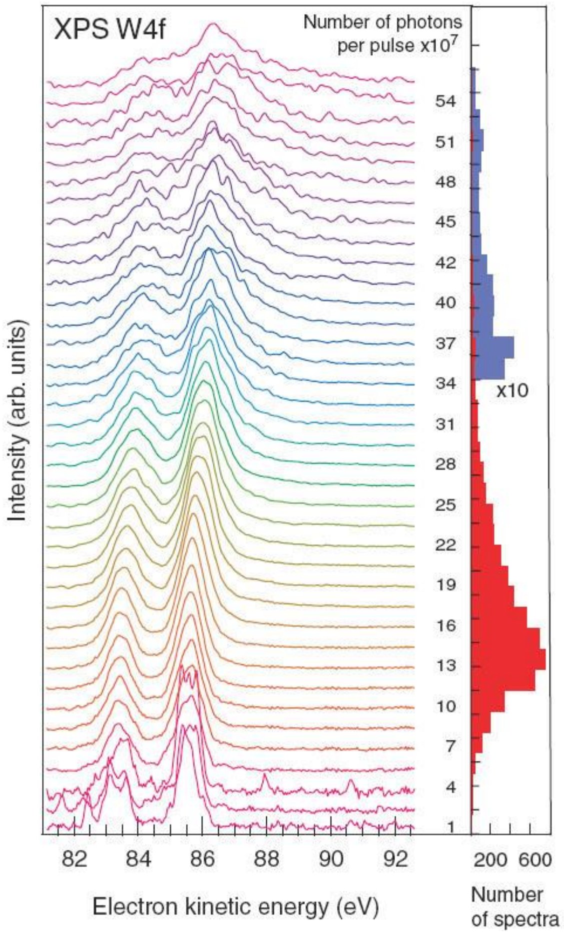


R. Cucini et al et al., NIMA (2011),
F. Bencivenga, JSR 23 (2016)

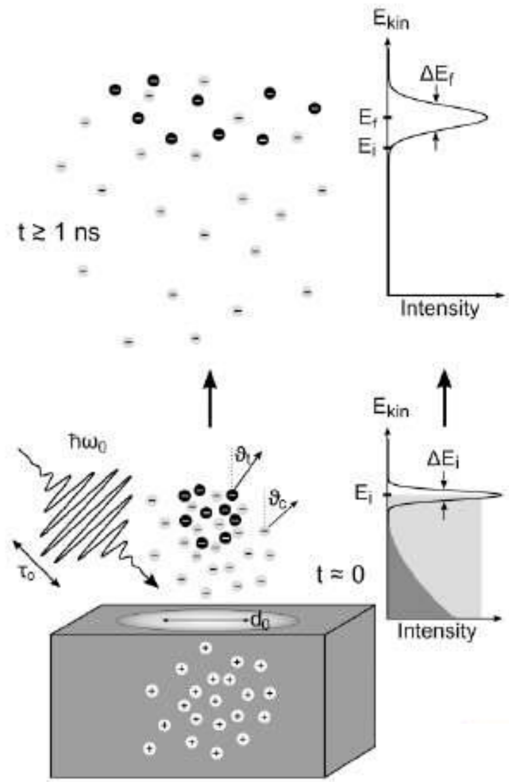
Elettra Sincrotrone Trieste

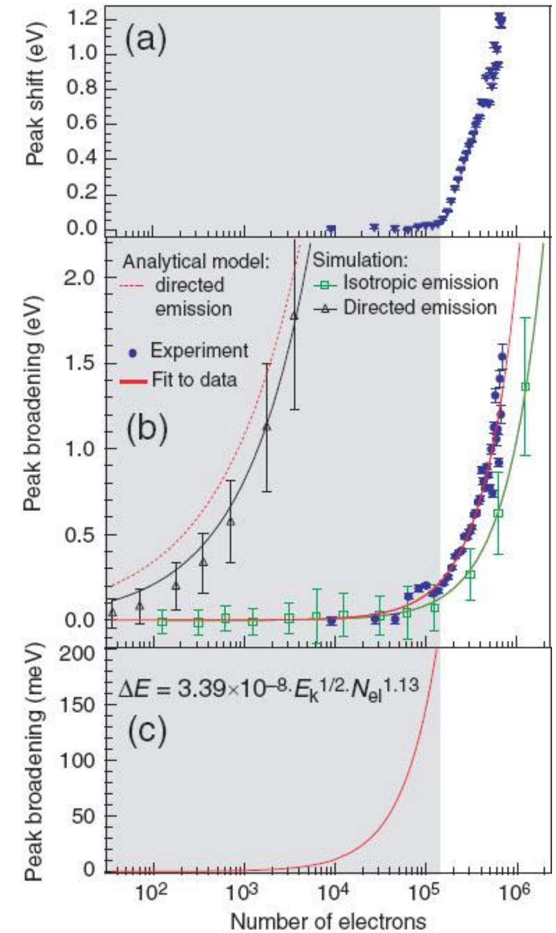
PES with FELs???

Core-level PE was proven to be extremely useful tool for time-resolved studies but FELs are too bright!.....Hrep_XFEL just started



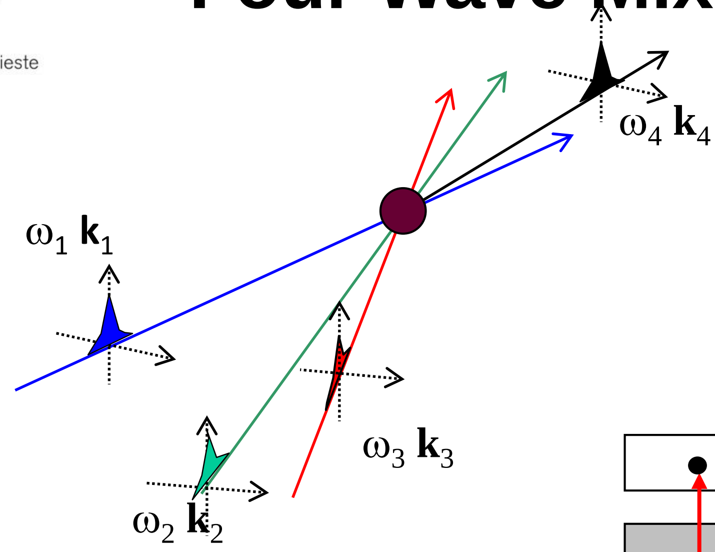
space charge (1mm spot)
> 10^8 phot/pulse
> 10^4 el/pulse



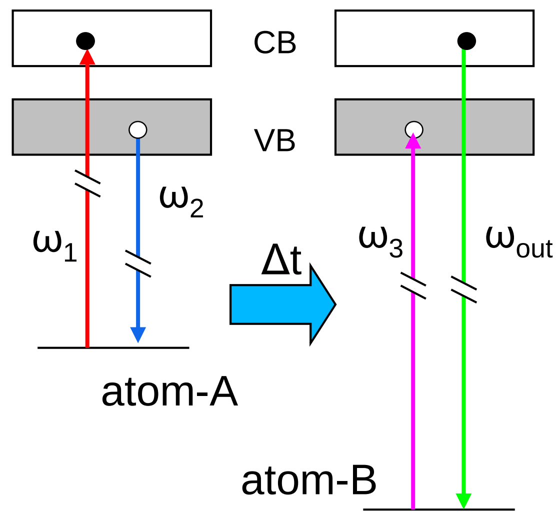


Momentum-Time resolved resonant inelastic x-ray scattering with high spectral resolution is feasible and complementary.

Four Wave Mixing at FEL's what is next



Transient grating is one of **Four Wave Mixing** techniques
Coherent Antistokes Raman Scattering (**CARS**)



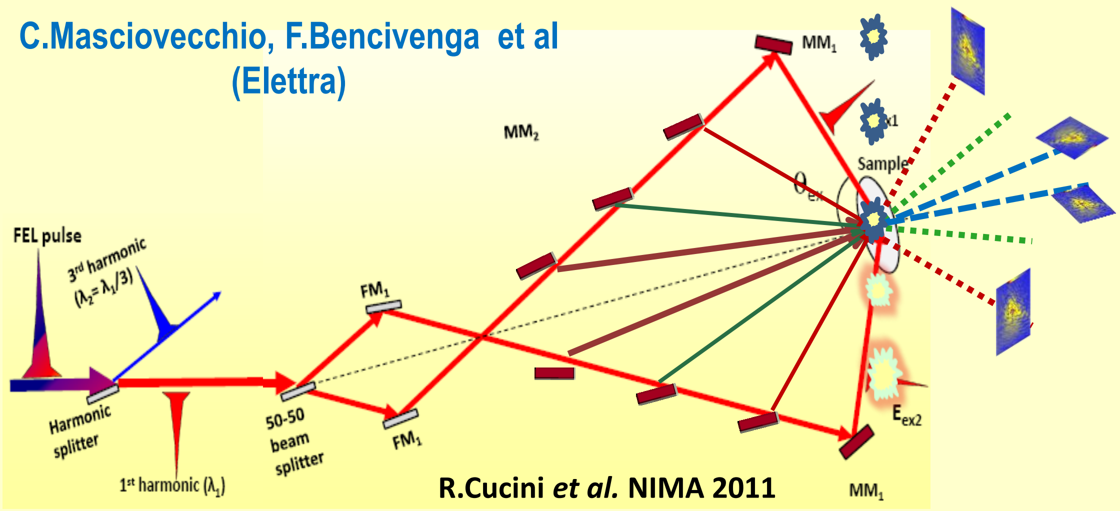
S. Tanaka & S. Mukamel PRL (2002)

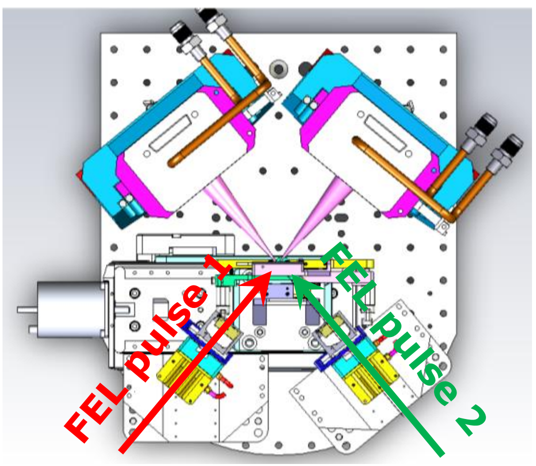
Measure the coherence between the two different sites → it makes possible to chose where a given excitation is created, as well as where and when it is probed

⬇

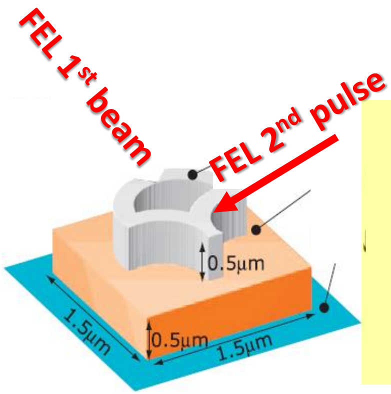
delocalization of electronic states and charge/energy transfer processes

Future developments (TIMER and stero/strobo-CDI)

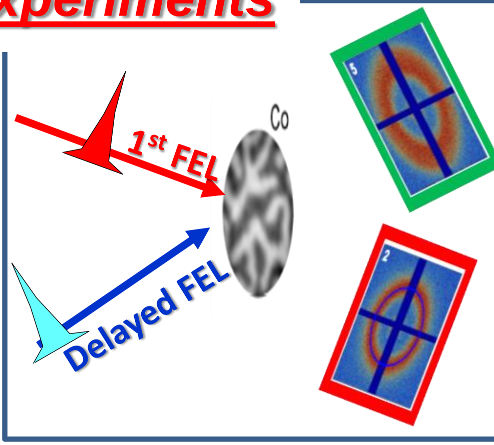




Possible experiments



Different view angles reveal different features, easier to reconstruct and perform a stereo-CDI



Res-CDI,
Strobo FEL-FEL P&P experiment recorded by different CCD.
Strobo-CDI

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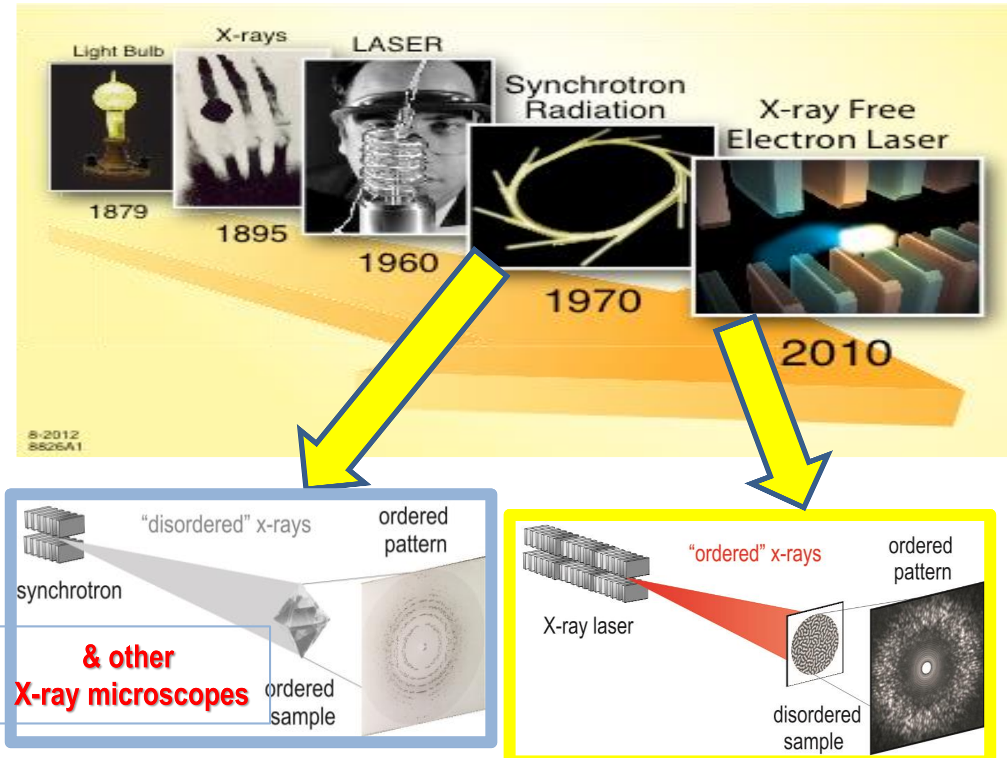
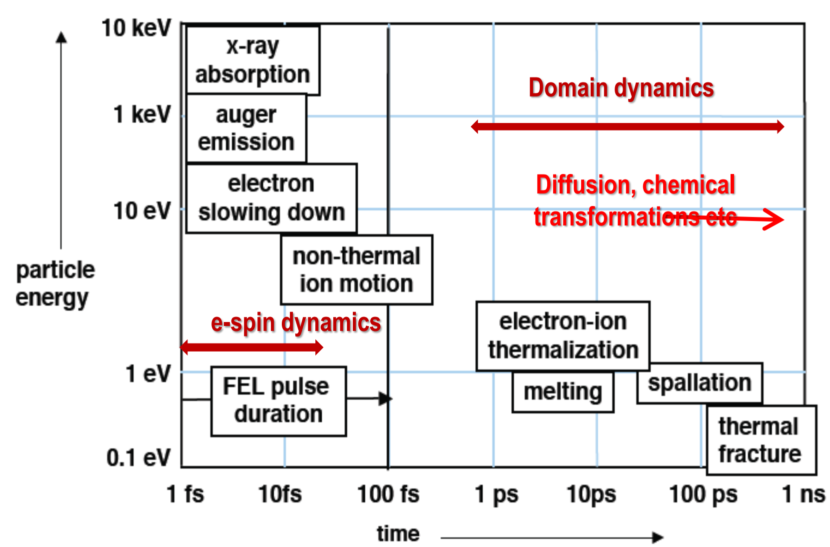
X-ray sources complementary used in material science: from static to dynamics



Energy-time picture of x-ray material interaction



- ❖ Micro-nanoscale order
- ❖ Equilibrium States
- ❖ Slower processes > ps

new paradigm ➔

- ➢ Nanoscale order
- ➢ Fast dynamics < ps
- ➢ Excited transient states

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